Nonlinear $\pi$ phase shift for single fibre-guided photons interacting with a single resonator-enhanced atom

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Realizing a strong interaction between individual photons is an important objective of research in quantum science and technology. It requires an optical medium in which light experiences a phase shift that depends nonlinearly on the photon number. Once the additional two-photon phase shift reaches $\pi$, such an ultra-strong nonlinearity could enable the implementation of high-fidelity quantum logic operations. However, the nonlinear response of standard optical media is orders of magnitude too weak. Here, we demonstrate a fibre-based nonlinearity that realizes an additional two-photon phase shift close to the ideal value of $\pi$. We employ a whispering-gallery-mode resonator, interfaced by an optical nanofibre, where the presence of a single rubidium atom in the resonator mode results in a strongly nonlinear response. We show that this results in entanglement of initially uncorrelated incident photons. This demonstration of a fibre-integrated, ultra-strong nonlinearity is a decisive step towards photon-based scalable quantum logics.

Optical photons are a key ingredient for investigations and applications in modern quantum information science. They are well decoupled from their environment but can, nevertheless, be conveniently manipulated with high precision. In conjunction with the ease of transmitting them over long distances using optical fibres, this makes photons prime candidates for the distribution and processing of quantum information in scalable quantum networks as well as for metrology beyond the standard quantum limit. These and many other applications require the photons to interact with each other in order to prepare and probe entanglement or to perform quantum logic operations. However, a direct photon–photon interaction does not exist in free space. It has been shown that an effective photon–photon interaction can be implemented probabilistically using linear optical elements in combination with projective measurements. Yet, for now, this technique cannot be considered scalable in a practical sense because it requires high-fidelity single-photon sources and detectors that are still beyond present capabilities. Alternatively, a deterministic interaction can be realized by means of an optical medium that exhibits a nonlinearity down to the level of individual photons. Such strong optical nonlinearities have been demonstrated, for example, in atomic ensembles, where the nonlinearity either stems from direct interaction between the excited atoms or from the generation of an intrinsic Kerr nonlinearity via electromagnetically induced transparency. An alternative approach is based on enhancing the nonlinearity of individual quantum emitters by coupling them to optical resonators. With these systems, nonlinear phase shifts up to a few tens of degrees, single-photon-controlled on- and off-switching of light, as well as photon number-dependent redirection of light have been demonstrated. In this Article, we demonstrate the realization of an optical fibre-based nonlinearity that leads to an unprecedented two-photon phase shift close to the ideal value of $\pi$, following a proposal by Hofmann and co-authors.

Principle
The nonlinearity is induced by a single $^{85}$Rb atom that is coupled to a whispering-gallery-mode (WGM) resonator while the latter is efficiently interfaced by an optical nanofibre. Our experimental set-up is sketched in Fig. 1. The central element is a so-called bottle microresonator, a novel type of WGM microresonator that is conceptually similar to other WGM microresonators but has the additional advantage of being fully tunable. The resonator is interfaced with a tapered fibre coupler that has a nanofibre waist. Resonant, horizontally ($H$) polarized light that is guided in the nanofibre will partially couple into the empty resonator and then couple back into the fibre, thereby acquiring a phase shift of $\pi$ due to the interaction with the resonator mode. The distance between the nanofibre and the resonator is chosen such that the system operates in the overcoupled regime; that is, the coupling rate $\kappa$ between the nanofibre and the resonator is larger than the intrinsic resonator loss rate $\kappa$. In this regime, the amplitude of the light that couples from the resonator back into the fibre is larger than that of the light transmitted past the resonator, and the phase of the total light field in the nanofibre after interaction with the resonator also equals $\pi$. The situation is different if the photon is vertically ($V$) polarized or if an atom is coupled to the resonator. In the former case, the light cannot enter due to the large birefringence of the resonator, whereas in the latter case, the presence of the atom significantly suppresses the intra-resonator field and thus changes the effective resonator loss rate (see Methods), which puts the system in the undercoupled regime. In both cases, the photons that arrive one by one therefore do not acquire a phase shift. However, in our scheme, the atom acts as a saturable absorber. In particular, the atom-induced losses per photon are significantly smaller when two $H$ photons are incident on the resonator at the same time. In this situation, the system is again in the overcoupled regime and the two-photon wavefunction acquires a phase $\pi$.

This can be measured experimentally by comparing the $H$-polarized light with the uncoupled $V$-polarized light field, which acts as a phase reference. Neglecting optical losses and choosing an input polarization along the +45° (that is, $H+V$) direction, the...
quantum state of the two photons before and after interaction with the atom–resonator system is given by

\[
\psi_{\text{initial}} = \frac{1}{\sqrt{2}} \left( a_{1+}^\dagger a_{1H}^\dagger + 2a_{1+}^\dagger a_{1V}^\dagger + a_{2+}^\dagger a_{2D}^\dagger \right) |0\rangle = \frac{1}{\sqrt{2}} a_{D+}^\dagger a_{D}^\dagger |0\rangle
\]

\[
\psi_{\text{final}} = \frac{1}{2\sqrt{2}} \left( -a_{1+}^\dagger a_{1H}^\dagger + 2a_{1+}^\dagger a_{1V}^\dagger + a_{2+}^\dagger a_{2D}^\dagger \right) |0\rangle
\]

\[
= \frac{1}{2} \left( a_{V+}^\dagger a_{V}^\dagger - a_{D+}^\dagger a_{D}^\dagger \right) |0\rangle
\]

respectively. Here, we have introduced the creation operators for horizontal, vertical, plus (D) and minus (D) 45°-polarized photons: \( a_{1+}^\dagger, a_{1V}^\dagger, a_{D+}^\dagger = (a_{1+}^\dagger + a_{1V}^\dagger) / \sqrt{2} \) and \( a_{D-}^\dagger = (a_{1+}^\dagger - a_{1V}^\dagger) / \sqrt{2} \), respectively. Due to the nonlinear interaction, an additional phase occurs for the \(|HH\rangle\) term, which, in the case of a \(\pi\) phase shift, results in the minus sign in equation (1). As a consequence, the final state is no longer separable. In the experiment, optical losses, fluctuating detunings and birefringence will inevitably change the probabilities for the outcome of the individual states and lead to decoherence of their relative phase. One thus has to verify the fidelity with which the nonlinear interaction leads to the ideal output \(|\psi_{\text{final}}\rangle\) and if the entanglement is indeed prepared under realistic conditions.

**Experimental procedure**

To optimize our atom–resonator system, we first analysed its performance at the single photon level before investigating the nonlinear phase shift. For this purpose, we tuned the bottle resonator and the incident light into resonance with the \(F = 3 \rightarrow F = 4\) \(^{85}\)Rb transition of a transverse magnetic (TM)-polarized resonator mode. In this case, the electric field has a dominant component perpendicular to the resonator surface, and the strong lateral confinement of the resonator mode gives rise to a strong longitudinal field component in the evanescent field. This component oscillates in quadrature with the transversal component and, as a consequence, the evanescent field of the clockwise-propagating light is nearly perfectly circularly (\(\sigma^+\)) polarized, where the quantization axis points along the resonator axis. Due to time reversal symmetry, the counterpropagating resonator mode is nearly perfectly \(\sigma^-\) polarized. The interaction with the clockwise-propagating resonator light field optically pumps the atom into the hyperfine ground state \(F = 3, m_F = 3\), and subsequently drives the cycling transition \(F = 3, m_F = 3 \rightarrow F = 4, m_F = 4\). In this case, the atom cannot emit photons into the empty, counterpropagating, \(\sigma^+\) polarized resonator mode. Thus, the atom only interacts with a single resonator mode, similar to the situation in a strongly birefringent, single-sided Fabry–Perot resonator.
Single photon polarization change.

Overlap of the polarization of the transmitted light with D-polarization and survival probability of the incident photons as a function of the resonator-fibre coupling strength $\kappa_D$ (in units of the intrinsic resonator loss rate $\kappa_s = 2\pi \times 8.4$ MHz). The input polarization was set such that, without an atom coupled to the resonator, the transmitted light was fully D-polarized. Error bars correspond to the 1σ statistical error, and solid lines are theoretical predictions that take into account a variation of the atom–resonator coupling strength and a residual atom–resonator and atom–light detuning of $\Delta_\sigma = \Delta_\rho = 2\pi \times 2.2$ MHz. The maximum overlap with D-polarization is observed at $\kappa_D = 2\pi \times 17$ MHz, in good agreement with the theoretical prediction (solid lines). The dashed line indicates the working point for the Kerr nonlinearity $\kappa_{\text{sat}} = 2.8\kappa_D$.

In the present experiment we adjusted the ratio of the amplitudes of the two polarization components, $H$ and $V$, of the incident light field such that they had the same amplitude after interaction with the empty resonator. Without an atom, the resonator imprints a phase shift close to $\pi$ on the $H$ component of the incident light and switches its polarization to $D$, thereby minimizing the power at the $D$-detector in the case when no atom is coupled. When an atom couples to the resonator, this situation changes and the phase shift disappears. This results in a measurable change in the polarization of the transmitted light. To couple atoms to the resonator we used an extension of the technique present by Junge et al., which allows atoms to be detected in the overcoupled regime where the fibre transmission is only marginally affected by the presence of the atom. An atomic fountain delivers a cloud of around $5 \times 10^7$ laser-cooled atoms to the resonator. We used a field-programmable gate array (FPGA)-based real-time detection and control system that permanently monitors the count rate of the $D$-detector. When an atom enters the resonator mode, it changes the polarization of the light field transmitted through the nanofibre and the count rate increases by two orders of magnitude. The FPGA detects this increase within $\sim 150$ ns. We then collected our measurement data in the subsequent 500 ns time interval. Following data acquisition we probed the resonator for 1 $\mu$s, which ensured that the atom was still coupled to the resonator mode at the end of the measurement sequence.

Linear phase shift

We first analysed the polarization change of the transmitted light between the empty resonator case and the case where an atom is coupled as a function of the fibre–resonator coupling strength $\kappa_D$. The latter is adjusted by changing the resonator–nanofibre distance. From a polarization analysis, we determined the overlap of the output light field with D-polarization as well as the survival probability of the incident photons (Supplementary Section I). The results are shown in Fig. 2. One can observe a monotonous increase in the photon survival probability with coupling rate $\kappa_D$ while the overlap with D-polarization reaches a maximum of 0.82 for $\kappa_D = 2\pi \times 17$ MHz, in good agreement with the theoretical prediction (solid lines in Fig. 2). To model this behaviour, we assumed a normal distribution of the atom–resonator coupling strength $g$, which originates from the motion of the atom through the resonator mode. From the transmission properties of our atom–resonator system, we calculated the corresponding polarization change (see Methods). We then fit this distribution to the measured data, with the mean coupling strength $\langle g \rangle$, and the standard deviation, $\sigma_g$, as the only free parameters. From this fit we obtain $g = 2\pi \times (13.5 \pm 1.5)$ MHz and $\sigma_g = 2\pi \times 4$ MHz.

Nonlinear phase shift

We choose the working point $\kappa_{\text{sat}} = 2.8\kappa_D = 2\pi \times 23.5$ MHz (see dashed line in Fig. 2) at which we expect photon number-independent loss and, as a consequence, an effectively dispersive nonlinearity (see Methods). At this point, the atom–resonator system yields a polarization-dependent transmission between 20% for $H$- and close to 100% for $V$-polarization. To experimentally determine the nonlinear phase shift, we modelled the transmitted light in three complementary polarization bases by recording coincidence counts between the different detectors (Fig. 1). Figure 3 exemplarily shows the measured coincidences recorded for the combinations $R/L$, $R/D$, $D/D$ and $D/H$. We clearly observe photon bunching and anti-bunching for zero time delay that vanishes when the time delay between the photons increases beyond the effective atomic lifetime $\tau_{\text{sp}} = 8$ ns. Here $\tau_{\text{sp}} = 1/2\Gamma$, with the Purcell-enhanced emission rate $\Gamma = g^2/(\kappa_s + \kappa_D) + \gamma$ (ref. 16). The (anti-)bunching indicates that two simultaneously arriving photons have a different polarization from individual photons.

Results and discussion

From the set of independent tomographic measurements we then reconstructed the density matrix of the two-photon state after interaction with the resonator using a maximum likelihood estimation. Figure 4e shows the real and imaginary parts of this reconstructed density matrix $\rho_{\text{final}}$, for zero mean detection time difference and a width of 3 ns for the coincidence window, which is in good qualitative agreement with the ideal density matrix $\rho_{\text{final}} = \ket{\psi_{\text{final}}}\bra{\psi_{\text{final}}}$ shown in Fig. 4f. For comparison, we also recorded the correlations without an atom coupled to the resonator. There, no significant bunching or anti-bunching is apparent and the reconstructed density matrix corresponds, as expected, to the separable state $\rho_{\text{sp}} = 1/2 (\rho_{\text{H}} + \rho_{\text{V}})$ (Supplementary Section III). This confirms that the atom is the physical origin of the nonlinearity.

The purity of the final state $\rho_{\text{final}}$ is 0.48 ± 0.02, indicating a significant reduction of coherence. This originates predominantly from averaging over shot-to-shot fluctuations of the atomic position, which results in a fluctuating atom–resonator coupling strength and a concomitant variation of the output state. Another source of decoherence originates from cases where three photons simultaneously interact with the resonator, which we cannot distinguish from two-photon states. For our experimental setting, the average photon number of the incident light within the 3 ns coincidence window is 0.04, from which, together with the known losses and detection efficiencies, we estimate that ~4.1% of the detected coincidences originate from three-photon events. Figure 4e also shows the overlap of the measured two-photon state with the ideal state, $\bra{\psi_{\text{final}}}\rho_{\text{final}}\ket{\psi_{\text{final}}}$, as well as the nonlinear phase shift as a function of the mean delay between the detections of the two photons and of the width of the coincidence window. We find a maximum overlap of 0.57 ± 0.02 and a nonlinear phase shift of $(1.05 \pm 0.04)\pi$, close to the value of $\pi$ expected under ideal conditions.

As is apparent from equation (1), the $\pi$ nonlinear phase shift ideally generates a non-separable state of light from the initially uncorrelated photons. To quantify the non-classical character of the experimentally prepared two-photon state, we calculated its concurrence (see Methods). We found a maximum concurrence of...
Figure 3 | Tomographic data. Measured coincidence counts between different detectors in Fig. 1 as a function of the delay between detection events (bin size = 1 ns) for a total measurement time of 28 h. Data are corrected for optical losses to the respective detectors (see Methods and Supplementary Section II). The case where an atom couples to the resonator is shown by blue data. For comparison, the empty resonator case is shown in red. For clarity, we exemplarily show only the correlation functions between the detection bases $R/L$, $R/D$, $D/D$ and $H/D$, which exhibit clear photon bunching and anti-bunching. The full set of measurements, including other combinations of bases, is presented in Supplementary Section II.

Figure 4 | State tomography of transmitted light. a, Overlap of the experimentally prepared state with the ideal state $\rho_{\text{final}}$. b,c, Concurrence (b) and nonlinear phase shift (c) of the two-photon state as a function of the coincidence window size and the mean photon–photon delay. Data are calculated from the corresponding density matrices, which are obtained from a maximum likelihood estimation. d, Overlap, concurrence and nonlinear phase shift as a function of mean photon–photon delay for the lines indicated in a–c. For reference, the concurrence in the case where no atom couples to the resonator is also shown. Error bars were determined by adding Poissonian noise to the measured coincidences, followed by a density matrix reconstruction and subsequent evaluation of the respective quantity. For each data point we generated a set of 100 random density matrices and used the resulting standard deviation as our error estimate. e,f, Real (red) and imaginary (blue) parts of the reconstructed density matrix $\rho_{\text{meas}}$ for zero time delay (e) and of the density matrix $\rho_{\text{final}}$ of the ideal state (f).
0.27 ± 0.04 at zero mean detection time difference, thereby clearly demonstrating that $P_{\text{meas}}$ corresponds to an entangled state. The concurrence and, thus, the entanglement vanishes when the time delay between the photons increases beyond the effective atomic lifetime $\tau_{\text{eff}} = 8$ ns (Fig. 4). This is expected, because for larger delays the photons interact one by one with the atom–resonator system.

**Outlook**

In summary, we have demonstrated an ultra-strong optical nonlinearity that leads to a nonlinear $\pi$ phase shift for coincident photons. The employed experimental platform is fully fibre-integrated and therefore compatible with optical fibre communication networks. The performance of the resonator can be improved by increasing the photon lifetime in the resonator and by trapping the atom in a small volume close to the resonator, thereby avoiding the variation of the atom–resonator coupling strength. For a state-of-the-art fibre microresonator\(^{19}\), this would result in a transmission in excess of 82%. When interfacing the atom–resonator system with two coupling fibres in an add–drop configuration, the nonlinearity can be exploited for the implementation of photon number-dependent routing\(^{21}\). Moreover, it would allow one to implement an efficient, error-proof Bell state analyser\(^{17}\) and thereby provide an essential ingredient in quantum communication and optical quantum information processing\(^{4}\). An optical nonlinearity that is generated by a two-level atom does not allow one to directly implement a deterministic two-photon quantum gate\(^{26,29}\). However, the single rubidium atom in our scheme can be straightforwardly prepared in a quantum mechanical superposition of its two hyperfine ground states, of which only one is coupled to the resonator mode\(^{20-23}\). Together with the demonstrated conditional $\pi$ phase shift, this is the basis for realizing a single-photon transistor\(^{24}\) as well as a quantum-controlled phase-flip gate\(^{24}\), which would then enable deterministic quantum computation protocols with photons.

**Methods**

**Single-photon phase shift.** Due to the birefringence of the resonator, V-polarized light does not couple to the resonator and the complex amplitude transmission $t_V$ through the coupling fibre is unity. In contrast, H-polarized light always couples to the resonator and the transmission is given by

$$t_H = \frac{\kappa_V - \kappa_H + i\Delta}{\kappa_V + \kappa_H + i\Delta} \tag{2}$$

Here, $\Delta$ is the fibre–resonator coupling strength, $\kappa_H$ the resonator loss rate and $\Delta$ the detuning between the resonator and the light field. This expression allows us to treat both cases—the empty resonator and the coupled atom–resonator system—by introducing two loss rates. For the empty resonator, the loss rate is given by the intrinsic loss rate, $\kappa_V = 2\pi \times 6.4$ MHz, while for the coupled atom–resonator system in the single-photon limit, the losses are governed by an effective loss rate that also depends on the atom:

$$\kappa_H = \frac{\gamma^2}{g^2 + i\Delta} + \kappa_V$$

Here, $\gamma$ is the atomic decay rate, $g$ is the atom–resonator coupling strength, and $\Delta$ is the detuning between the atom and the light field. Using these expressions, we model the phase shift and transmission properties of the resonator and calculate the expected polarization change of the transmitted light. For the case where both atom and resonator are resonant with the incident light field ($\Delta = \Delta = 0$), equation (2) predicts a phase shift of 0 and $\pi$ for the undercoupled ($\kappa_V > \kappa_H$) and overcoupled ($\kappa_V < \kappa_H$) regime, respectively.

**Atom–resonator interaction beyond the single-photon limit.** To simulate the behaviour of the atom–resonator system for the case with more than one photon present, we performed a full numerical calculation of our system by solving the master equations of the pumped atom–resonator system using the approximation of a two-level atom interacting with a single resonator mode\(^{21}\). In this case, the master equation is given by

$$\frac{\text{d} \rho(t)}{\text{d}t} = -i[H, \rho] + (\kappa_V + \kappa_H)2|b\rangle\langle b| - |b\rangle\langle b| + \frac{1}{2}(2g^2 - b^\dagger b - b\dagger b + b^\dagger b) + \sqrt{2\Delta}(\rho_{\text{H}}(b^\dagger b + b)$$

where $H / \hbar = \Delta_0 |b\rangle\langle b| + \Delta |a\rangle\langle a| + g(|b\rangle\langle a| + b^\dagger a^\dagger) + \sqrt{2\Delta}(\rho_{\text{H}}(b^\dagger b + b) + \rho_{\text{V}}(b^\dagger b + b^\dagger(b\dagger b + b))$

is the atom–resonator Hamiltonian in the rotating wave approximation and $b (b^\dagger)$ is the annihilation (creation) operator of a resonator photon, $a^\dagger (a)$ is the atomic excitation (de-excitation) operator, $\rho$ is the atom–resonator density matrix, and $(\rho_{\text{H}})$ is the amplitude of the incident $H$-polarized fibre guided light. The final state of the light field after interaction with the resonator is given by the interference of the incident field with the field that is coupled out of the resonator:

$$\rho = a_{\text{H}}(b^\dagger b + b)$$

The input light field corresponds to a coherent state, which allowed us to replace the input operators $a_{\text{H}}$ and $a_{\text{V}}$ by their expectation values\(^{25}\). Using the measured distribution of coupling strengths, we calculated the expectation values for the output fields, which allowed us to predict the optical losses in the system and to estimate the optimal working point, where the two-photon transmission is independent of the photon time delay.

**State reconstruction.** For the reconstruction of the density matrix of the two-photon state after the interaction with the resonator, we used the 19 non-trivial coincidence detections obtained from the polarization analyser set-up. We corrected the measured coincidence rate by the product of the single photon transmission probability for the respective detector pairs (see Supplementary Section II for details). Note that we did not record coincidences for the settings $RR$ and $VV$. We defined a coincidence window and summed up all two-photon events within this window. The incident photons originate from a coherent laser beam with a coherence time on the order of $1 \mu$s and are thus, to a very good approximation, indistinguishable within the interaction time, which is given by the inverse of the resonator increased emission rate $\tau_{\text{eff}} = 1/2\Gamma = 8$ ns. As a consequence, the two-photon state is limited to the symmetric subspace of a two-qubit Hilbert space. The corresponding basis states are $|HH\rangle$, $|VV\rangle = (|HV\rangle + |VH\rangle)/\sqrt{2}$ and $|VV\rangle$ and the density matrix can be written as

$$\rho = (\rho_{\text{HH}} \rho_{\text{RV}} \rho_{\text{HV}VV} \rho_{\text{RH}} \rho_{\text{PS} \rho_{\text{PV}} \rho_{\text{VV}}}) = (0 0 0 0 0 0 0 0 0)$$

The matrix element $\rho_{\text{PS}}$ and all other matrix elements that contain contributions from the asymmetric basis state $|A\rangle = (|HV\rangle - |VH\rangle)/\sqrt{2}$ and the 3 x 3 sub-matrix contains the full information on the experimentally prepared state. The 3 x 3 sub-matrix is reconstructed by performing a quantum state tomography using a maximum likelihood estimation\(^{26}\).

**Concurrence and nonlinear phase shift.** To quantify the degree of entanglement, we transformed the full 4 x 4 density matrix into an unentangled basis defined by the basis states $|HH\rangle$, $|VV\rangle$, $|HV\rangle$ and $|VV\rangle$ and subsequently calculated the concurrence\(^{27}\). The nonlinear phase shift was determined directly from the measured 3 x 3 density matrix according to $\phi_{\text{PS}} = \phi_{\text{HH}} - 2\phi_{\text{PS}} = \arg(\rho_{\text{HHV}}) - \arg(\rho_{\text{PVV}})$, where these two density matrix elements were chosen because of their higher signal-to-noise ratio.

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Author contributions
All authors contributed to the experiment, the analysis of the results and the writing of the manuscript.

Additional information
Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to J.V. and A.R.

Competing financial interests
The authors declare no competing financial interests.