Correlating photons using the collective nonlinear response of atoms weakly coupled to an optical mode

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Photons in a nonlinear medium can repel or attract each other, resulting in strongly correlated quantum many-body states2–5. Typically, such correlated states of light arise from the extreme nonlinearity granted by quantum emitters that are strongly coupled to a photonic mode6–7. However, unavoidable dissipation (such as photon loss) blurs nonlinear quantum effects when such approaches are used. Here, we generate strongly correlated photon states using only weak coupling and taking advantage of dissipation. An ensemble of non-interacting waveguide-coupled atoms induces correlations between simultaneously arriving photons through collectively enhanced nonlinear interactions. These correlated photons experience less dissipation than the uncorrelated ones. Depending on the number of atoms, we experimentally observe strong photon bunching or antibunching of the transmitted light. This realization of a collectively enhanced nonlinearity may turn out to be transformational for quantum information science and opens new avenues for generating non-classical light, covering frequencies from the microwave to the X-ray regime.

Photons that strongly interact via a quantum nonlinear medium exhibit complex out-of-equilibrium quantum many-body dynamics that may enable one to tailor and control the photon statistics of the light8–10. The resulting quantum correlated light can then act as a key resource in quantum sensing, quantum metrology and quantum communication, as well as quantum simulations and information processing. Recently, important advances have been made in mediating interactions between optical photons by strongly coupling them to quantum emitters and exploiting the inherently nonlinear response of the latter2–3. Several methods have been used for this purpose, such as resonant enhancement via high-finesse optical cavities4–6, collective responses of strongly interacting Rydberg atoms11–18 or efficient coupling of single quantum emitters to waveguide39–42. However, the implementation of strong interactions between individual optical photons remains a challenge. In particular, such approaches are often impaired by unavoidable dissipative processes that cause photon loss and blur nonlinear quantum effects. Here, we experimentally demonstrate a mechanism by which a strongly dissipative nonlinear medium that consists of non-interacting quantum emitters weakly coupled to a light field is harnessed to generate strongly correlated states of light43. Specifically, we launch a weak resonant laser light field through an ensemble of atoms. As each atom can absorb only one photon at a time, photons arriving at the same time experience an effective interaction, which is witnessed through a modification of the second-order correlation function, \( g^{(2)}(r) \), of the light, where \( r \) is the time difference. Adjusting the number of atoms, we continuously change the photon statistics from antibunching down to \( g^{(2)}(0) = 0.37 \pm 0.12 \) to bunching of up to \( g^{(2)}(0) = 24 \pm 7 \). This demonstrates coherent collective enhancement of photon–photon interactions in an ensemble of otherwise non-interacting emitters. Consequently, our scheme may be transformational in quantum information science. For example, it offers a new approach to realizing single-photon sources which may outperform sources based on single quantum emitters with comparable coupling strength.

The experimental set-up is shown in Fig. 1a. An ensemble of laser-cooled caesium (Cs) atoms is trapped along a 400-nm-diameter optical nanofibre, which is realized as the waist of a tapered optical fibre. The trapping potential consists of two diametric linear arrays of individual trapping sites located at a distance of ~250 nm from the fibre surface. Each site contains at most one atom and offers subwavelength confinement of the atoms in all three spatial dimensions44. The lattice spacing is not commensurate with the wavelength of the guided probe light and the filling factor of the trapping sites is about 0.1. Under these conditions, the response of the atomic array is effectively the same as that of an unordered medium (see Methods). We send probe light through the nanofibre that couples to the atoms via the evanescent field and is resonant with the cycling transition of the Cs D2 line (6S\(_{1/2}\), \( F = 4 \rightarrow 6p_{3/2}\), \( F = 5 \), wavelength \( \lambda = 52 \) nm). The input power of the light is \( P_0 = 2.35 \) pW, which corresponds to a saturation parameter of \( S_0 = P_0 / P_{sat} = 0.02 \) where \( P_{sat} \) is the power required to obtain saturation intensity at the first trapping site. A bandpass filter at the output is used to remove spurious, trap light-induced Raman fluorescence from the transmitted light. The probe field is quasi-linearly polarized45, and the polarization axis is aligned such that the light exhibits near-circular polarization at the position of the atom. This realizes chiral light–matter coupling46, whereby the atoms predominantly interact with light propagating in the forward direction and backscattering is strongly suppressed. The coupling strength of the atoms to the forward propagating nanofibre mode, \( \beta = F / \Gamma \), is defined as the ratio of the spontaneous emission rate of an atom in the forward

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direction of the waveguide, $I_{\text{rat}}$, and the total spontaneous emission rate into all channels, $I_G$.

In the linear optical regime, the atoms act as a narrow-band spectral filter and induce a strong attenuation of individually propagating resonant probe photons that increases exponentially with the number of atoms. However, when two probe photons are incident with a time delay shorter than the excited state lifetime, the atom can absorb only one of the photons. In combination with stimulated emission, this process can be depicted as a nonlinear four-wave mixing process that induces energy–time entanglement between the photons. As a result, each of the two scattered photons then features a broadened spectrum with frequency components that are red- and blue-detuned from the atomic resonance (see Fig. 1b). The scattered photons thus experience reduced (subexponential) propagation loss\(^{23}\). The amplitudes of the correlated photon pairs that arise from scattering by individual atoms add up coherently, thus giving rise to a collective enhancement of the process. Eventually, the ratio of the unscattered and scattered two-photon components in the output mode is defined by the total number of atoms coupled to the optical mode, the strength of the two-photon scattering process and the linear loss of the two-photon component due to photon scattering out of the optical mode.

We measure the photon statistics of the transmitted light by sending it onto a Hanbury–Brown–Twiss set-up consisting of a 50/50 beam splitter, a single-photon counting module (SPCM) in each output and a time-tagging unit to record the photon arrival times (Fig. 1). The normalized histogram of the time differences between the photon detection events then yields $g^{(2)}(\tau)$ of the transmitted light (see Methods). In addition, we use the measured count rates from the SPCMs to determine the transmitted power through the nanofibre. This allows us to infer the optical depth (OD) of the trapped ensemble for each of the $2.6 \times 10^5$ experimental runs. After sorting the data according to the measured OD and averaging the data in each OD interval, we obtain a set of 54 second-order correlation functions; see Extended Data Fig. 1.

Figures 2 and 3 summarize the main results of these correlation measurements: Fig. 2 shows four examples of second-order correlation functions for different regimes and Fig. 3 shows the value of $g^{(2)}(\tau = 0)$ as a function of the OD for all measured correlation functions. The solid orange curves in both figures are theory predictions\(^{13}\) taking into account the experimental variation in the atom number distribution. The assumed coupling strength is derived from fitting the model to the data in Fig. 3 with $\beta$ as the only fit parameter; see Methods. The fitted value of $\beta = 0.82 \pm 0.02\%$ agrees well with the value of $\beta = 0.83 \pm 0.03\%$, derived from an independent saturation measurement; see Methods. The resulting theory curves also agree well with the experimental data in Figs. 2 and 3 and Extended Data Fig. 1. The fitted value of $\beta$ is also used to determine the number of trapped atoms from the measured OD.

For vanishing ODs, we observe a flat $g^{(2)}(\tau)$ with $g^{(2)}(0) \approx 1$, as expected for the photon statistics of our probe laser. With increasing OD, one observes photon antibunching—that is, $g^{(2)}(0)$ starts to fall below 1; see Fig. 2a. It originates from destructive quantum interference of the unscattered two-photon component and the scattered two-photon component. When a single resonant photon is absorbed and re-emitted, it acquires a phase shift of $\pi$. Similarly, when two photons are incident with a large time delay, both can
be absorbed and re-emitted, giving rise to a total phase shift of $2\pi$ of the scattered two-photon wave. However, if two photons arrive within a time interval that is small compared with the atomic lifetime, the scattered two-photon wavefunction only acquires a phase shift of $\pi$, as at any given time an atom can absorb and re-emit only one photon (see Methods). As a result, in addition to linear absorption of the two-photon component, its overall probability amplitude at zero delay is further reduced by destructive interference, which then leads to a reduction of $g^{(2)}(0)$ (see Methods). With increasing atom number, the two-photon components scattered by the individual atoms add up coherently, resulting in a more pronounced destructive interference and photon antibunching. For an OD of 5.13, corresponding to a mean number of atoms of $N = 156$, the antibunching reaches its smallest value of $g^{(2)}(0) = 0.37 \pm 0.12$ (Fig. 2b). Ideally, the theory even predicts perfect antibunching when the ratio of scattered to unscattered two-photon amplitudes is similar; see the dashed line in Fig. 3. The reduced contrast of the measured correlation functions stems largely from the fact that each correlation function is averaged over a spread of ODs. This is due to both the finite binning of ODs and photon shot noise impairing the precise determination of the OD from the transmission in the individual experimental runs.

When the OD or the number of trapped atoms are increased further, the ratio of scattered to unscattered photon pairs also increases. As a consequence, $g^{(2)}(\tau)$ starts to exhibit a peak at $\tau = 0$, and perfect anticorrelations for finite time delays $\tau \neq 0$ are expected from the theory; see the orange line in Fig. 2c. This predicted oscillatory behaviour of $g^{(2)}(\tau)$ originates from the quantum beat of the scattered and unscattered two-photon components\(^27\). For very large ODs, the scattered two-photon component dominates in the output field. This manifests in strong photon bunching, as observed in our measurements for mean atom numbers exceeding $N \approx 180$; see Figs. 2d and 3. In the extremely high OD limit, all transmitted photons originate from nonlinear interactions with the atoms and the bunching increases indefinitely.

We note that the setting and the underlying physics in our experiment are fundamentally different from experiments that study the photon statistics of atomic fluorescence\(^{21,22}\). The latter investigate light scattered into an optical mode that is separate from the incident light. As atoms can only scatter one photon at a given time, one observes maximum antibunching for a single atom, whereas when the number of atoms increases, $g^{(2)}(\tau = 0)$ also increases and the non-classical properties of the light decrease. In contrast, in our experiment we measure an almost flat $g^{(2)}(\tau)$ for a single atom. We observe sizeable photon antibunching only with substantially larger atom numbers. This behaviour reveals the collective nature underlying this process and shows that ensembles of weakly coupled atoms can be used to realize strongly correlated many-body states of photons. In contrast to previous approaches that rely on strong light–matter coupling and the suppression of dissipation, the underlying dynamics of our non-equilibrium many-body quantum system is based on an interplay of weak optical nonlinearities, collective enhancement and finite dissipation. It is well known that collective enhancement can be used to compensate for a limited coupling efficiency and enable strong coupling with ensembles of otherwise non-interacting atoms\(^{20,21}\). The generation of non-trivial field states have so far, however, relied on strong optical driving fields to enhance the optical nonlinearity. In contrast, no such driving fields are used in this experiment. The present approach thus extends the concept of collective enhancement to direct photon–photon interactions. This broadens the range of possible applications, particularly in the realm of quantum information science.

As an example, consider employing the observed antibunching for the generation of single photons. For small coupling strengths ($\beta < 0.1$) and low input photon rates ($n_{in} < 0.1I/\beta$), we can
approximate the power transmission in OD (7) at which we obtain perfect antibunching by $T \approx \beta$ (Methods). It is thus possible to realize a stream of antibunched light with a photon rate $n_{\text{rate}} \approx 0.1\,\text{f}^{-1}$ in the output mode, independent of the type of emitter and its coupling strength $\beta$ to the optical mode. Surprisingly, this is much larger than the maximum photon rate $\beta T / 2$ that is achievable with a single-photon source based on a single quantum emitter with the same coupling strength $\beta$. For sufficiently weak driving power, we expect these photons to be fully coherent and indistinguishable (Methods). In addition, this principle is independent of the type of emitter or specific optical mode or frequency used and can be achieved for all frequencies spanning the electromagnetic spectrum without the need for precise control of individual emitters and their coupling strengths. These features make the observed effects highly promising for realizing new sources of non-classical light, such as single-photon sources, particularly for wavelengths at which it is not possible to achieve strong coupling of individual atoms or emitters.

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Methods

Theoretical description of photon transport. The photon transport in this manuscript can be modelled by considering a weak coherent field with detuning $\Delta_0$ from atomic resonance and amplitude $a > 0$, which is truncated at two photons

$$|\text{in}\rangle \approx e^{-\alpha/2} \left( 1 + a^\dagger a + \frac{\alpha^2}{2} a^\dagger a a^\dagger a \right) |0\rangle.$$  \hspace{1cm} (1)

Here, $|0\rangle$ is the vacuum state with no photons in the waveguide and $a^\dagger a$ is an operator that creates a photon in the waveguide with detuning $\Delta_0$. We compute the output state by applying scattering matrices for $N$ two-level emitters on the one- and two-photon states. The scattering matrix is computed for a single-mode linearly dispersive waveguide with group velocity $v$ and unidirectional coupling.

The single-photon scattering matrix preserves the photon frequency, and after $N$ scattering events maps $a^\dagger \rightarrow a^\dagger \sum_i |\phi_i\rangle \langle \phi_i|$, where $t_j = 1 - 2\beta/(1 - 2\alpha I_j)$, where $I_j$ is the total decay rate of the two-level emitter. Owing to the unidirectional coupling, the distance between emitters amounts to only an overall phase and is ignored. Here, we consider resonant photons with $\Delta_0 = 0$ and therefore $t_{\text{out}} = 1 - 2\beta$. The single-photon part of the output state can thus be written

$$|\text{out}\rangle_1 = e^{-\alpha^2/2} \left( \sum_i |\phi_i\rangle \langle \phi_i| e^{-\alpha/2} \left( 1 + a^\dagger a \right) |0\rangle \right).$$

We compute two-photon transport using the procedure discussed in ref. 1. The two-photon part of the output state is computed in real space as

$$|\text{out}\rangle_2 = e^{-\alpha^2/2} \int dx_1 dx_2 \langle \psi(x_1, x_2) \phi(x_1) \phi(x_2) |0\rangle.$$  \hspace{1cm} (2)

Here the position coordinate is rescaled to give $x = x/v$. The expression for $\psi(x)$ highlights that it scales with the superposition of the exponentially damped term and the correlated term. Processes that scatter one photon from a two-photon input state out of the waveguide do not contribute to the leading-order expression of $\psi(x)$. Equation (2) is used to model the second-order correlation functions shown in the main text. We note that this model does not incorporate dephasing or other noise terms.

A general expression for $\phi_i(x)$ is cumbersome and can be found in ref. 1. Nevertheless, $\phi_i(x)$ takes on simple approximate forms in the small and large OD limits for a resonant drive, $\Delta_0 = 0$. In the small optical depth limit, $N\gg 1$, $\phi_i(x) \approx N \phi_i e^{-x^2/(2\sigma^2)}$. At the start of the ensemble for $N < 1/\beta$, $\phi_i$ grows linearly with atom number and the correlated transport is therefore collectively enhanced. For large optical depths, $N\gg 1$, the correlated part can be written as a Fourier integral of $\phi_i(x) \approx \int \frac{d\xi}{\sqrt{2\pi} \sigma} \cos(\Delta x/\sigma e^{-\xi^2/2}) e^{\xi x^2/2}$, where $\xi = N\phi(1-\beta)$. In this limit, the exponentially damped part vanishes and $\psi(x) \rightarrow \phi_i(x)$.

Coherence properties of the transmitted light. When light propagates through the ensemble, the latter acts as a photon-number-dependent filter. For individually propagating photons the medium's effect is the same as that of a standard linear filter. Thus it does not affect the coherence properties of the photons and the transmitted light retains coherence with the input light field. For a given atom number and sufficiently weak incident fields ($S_0 < 1$) the outgoing field is completely dominated by single-photon events and the nonlinear process only removes the few two-photon events. In this limit, the transmitted photons should be completely coherent with the incident field and indistinguishable from each other. As the saturation parameter increases, the nonlinearity contribution from two-photon processes will increase. As one approaches saturation intensity, for the point of perfect antibunching, these nonlinearly transmitted photons make up a significant fraction of all output photons. In this case, similar to the fluorescence of a strongly driven two-level emitter, the energy of the photons is in a mixed state with a frequency spread on the order of the atom's homogeneous width.

Depending on the desired application, the output photon rate must be balanced against the required purity and coherence of the output photons. For our experiment (saturation parameter $S_0 = 0.02$), we estimate for the point of best antibunching, the power from the individually propagating photons to exceed the nonlinear transmitted power by a factor of 45. Thus, the output photon stream is completely dominated by the single-photon component, and we expect it to be fully coherent with the input field, although we have not quantified this experimentally.

Nanofibre-based optical dipole trap. We trap laser-cooled Cs atoms using a nanofibre-based two-colour optical dipole trap30,31. The repulsive blue-detuned light field has a wavelength of $\lambda = 685$ nm and a power of $\sim 14$ mW, and is launched into the fibre in a running wave configuration. A pair of counter-propagating red-detuned fibre-guided light fields is also launched into the fibre, thereby forming a standing wave. This light forms an attractive potential and has a wavelength of $\lambda = 935$ nm and a power of $\sim 0.18$ mW per beam. All light fields are quasi-linearly polarized. The polarization planes of the two red-detuned light fields are parallel to each other, whereas the polarization plane of the blue-detuned transmitted light field is perpendicular to that. In this configuration, the minima of the optical trap potential are located at a distance of $\sim 230$ nm from the surface of the 400-nm-diameter nanofibre. The wavelengths of the trapping light fields correspond to the magic wavelengths of the Cs D2 line30, which minimizes the light shifts of this optical transition of the atoms in the nanofibre-based dipole trap. The distance between two adjacent minima of trapping sites along the axis of the nanofibre is 448 nm, which is non-commensurate with the probe wavelength. It is given by the wavelength of the red-detuned trapping light field and the effective refractive index of the nanofibre.

Ensemble preparation. The atoms are loaded into the optical dipole trap from a cigar-shaped cloud of cold Cs atoms. The atom cloud is created using a magneto-optical trap with elongated magnetic coils. The magneto-optical trap is followed by a molasses cooling stage in which the atoms are cooled down to sub-Doppler temperature and loaded into the trap. In our experiment, we reach typical filling factors of about 0.1, which we deduce from the spatial extension over which we load atoms into the nanofibre traps and the total number of trapped atoms. After the first molasses cooling stage, the magneto-optical and the magneto-optical trap magnetic fields are turned off and the red-detuned trap power is adiabatically ramped up over a time of 15 ms. This moves the potential minimum of the trap closer to the fibre surface and results in a larger coupling strength between the trapped atom and a nanofibre-guided light field. After the power ramp of the red-detuned light field, a second molasses phase cools the remaining trapped atoms. The OD exhibited by the trapped atoms is characterized by a standard frequency sweep. These steps are illustrated in Extended Data Fig. 2i–iv.

Probing sequence. The probe light field is resonant with the D2 $(6S_1/2, F = 4 \rightarrow 6P_{3/2}, F = 5)$ transition and launched as a travelling wave into the nanofibre. The probe two polarization is quasi-linearly polarized in the plane of the trapped atoms (that is, it coincides with that of the red-detuned trapping light fields). The probing sequence comprises 350 resonant pulses, each with a duration of 10 μs. The pulses have a power of 2.35 pW, which corresponds to a nanofibre saturation parameter $S_0 = P_{\text{in}}/P_{\text{sat}} = 0.02$ for the first atom the laser interacts with ($P_{\text{in}} = 50$ mW). The probe pulse duration is chosen such that the average kinetic energy that is transferred to the first atoms in the chain due to photon recoil is less than half the trap depth. To compensate for the heating, we apply molasses cooling with a duration of 200 μs between two probing pulses. The total number of probe pulses and the duration of the interleaved cooling pulses are chosen such that the change in OD over the entire sequence is as small as possible.

The probe sequence is followed by an additional frequency scan to measure the OD to check for atoms loss, see phase (vi) in the experimental sequence depicted in Extended Data Fig. 2. Afterwards, the atoms are removed from the trap, phase (vii), and we perform calibration measurements on the nanofibre transmission, phase (viii).

Detection. The light transmitted through the nanofibre-coupled atomic ensemble is first sent to a spectral filtering stage consisting of a Fabry–Perot cavity (spectral width: $\pm 100$ MHz) and a volume Bragg grating. In this way, the signal light can be separated from the trapping light fields and from the Raman scattering they produce when propagating in the fibre. The light is then sent onto a four-channel Brown–Twiss set-up that consists of a beam splitter with an SPCM in each output. The modules are connected to a field-programmable gate array (FPGA)-based time tagger that records the photon detection events with a timing resolution better than 1 ns. After plotting a histogram of the time differences between the photon detection events in the two detectors, we can infer the second-order correlation function of the transmitted light from the data:

$$S^{(2)}(\tau) = \left( \langle a^\dagger(t) a^\dagger(t+\tau) a(t+\tau) a(t) \rangle - \langle a^\dagger a^\dagger a a \rangle \right) / \langle a^\dagger a a \rangle.$$  \hspace{1cm} (3)

Here, $\langle a^\dagger a \rangle$ and $\langle a^\dagger \rangle$ are the photon annihilation and creation operators, respectively.

Data processing. The FPGA records time tags for each detection event in each SPCM. We aim to minimize the effect of transient transmission signals at the
beginning and end of each probe pulse, originating (for example) from the finite switching time of the acousto-optical modulator that controls the optical pulse. Thus, we only take detection events that fall into the probing interval between 1 μs and 9 μs for each probe pulse into account. Before each probe pulse train, at the beginning of each individual run, we also perform a measurement of the OD of the atomic ensemble. This increases the temperature of the atoms, and we observe an increased transmission for the first probe pulses that decays to a steady state after about ~15 probing–cooling iterations. To have a well-defined number of atoms and a precise atom–light coupling strength, we thus discard the data from the first 20 probe pulses of each experimental run.

From the timestamps recorded for a single run, we plot histograms of all of the time differences between the photon detection events in the two detectors to obtain the coincidence histogram $c(t)$, where $t$ represents the time delay between the two clicks. The bin size is 2 ns, and $c$ represents the number of coincidences detected at this time delay. We accumulate the coincidence statistics from each run into one out of 54 histograms based on the OD = −ln(T) observed in the individual run, where $T$ is the average transmission measured over the run. The resulting histograms are normalized by setting the correlation function to one for long time delays ($τ > 200$ ns). The OD windows used in Fig. 3 and in Extended Data Fig. 1 are chosen such that by increasing OD, the bin size also increases to compensate for the reduced signal to noise ratio in transmission. The windows correspond to OD bin sizes of 0.1, 0.2, 0.25 and 0.5 for ODs lying in the intervals [0, 4.4], [4.4, 5], [5, 6] and [6, 8], respectively.

Maximum-likelihood estimation of $g^{(2)}(0)$. To obtain a good estimate of the zero time-delay value $g^{(2)}(0)$ from the individual second-order correlation measurements, we perform a heuristic fit of the measured correlation functions in a small time window around $τ = 0$. Owing to the low coincidence count rates in the data for higher ODs, a standard fitting algorithm is in general not reliable. To circumvent this, we instead use a maximum-likelihood estimation method (MLE) that searches for the theory $g_{\text{theory}}^{(2)}(τ)$ function that has the highest probability of reproducing the measured correlation function. As the temporal distribution of the coincidences is directly proportional to the expected second-order correlation function $g_{\text{theory}}^{(2)}(r)$, this method boils down to maximizing the probability

$$L(θ) = \prod_{i=1}^{n} g_{\text{theory}}^{(2)}(r_i, θ)^{c(r_i)}$$

(4)

to observe the measured data assuming a theoretical correlation function $g_{\text{theory}}^{(2)}(r, θ)$ by varying the parameters of the set $θ$. Here, $c(r)$ represents the values of the measured coincidence histogram. As we are mainly interested in the zero value of the measured correlation functions, we can approximate them for small time delays by $g^{(2)}(τ) = 1 - A \exp(-Γ|τ|)$, where $A$ and $Γ$ are the two fit parameters. For optimizing the accuracy of the estimation of $g^{(2)}(0)$, we limit the fit region to the characteristic timescale of the expected correlation functions, that is, to $|Δτ| = 10 ns$ for the region where we observe antibunching and to $|Δτ| ≈ 15 ns$ for the datasets with OD > 6 where $g^{(2)}(0) > 1$ (Fig. 3).

Error estimation of $g^{(2)}(0)$. We estimate the error in the value of $g^{(2)}(0)$ by means of a bootstrapping method. For each data point in Fig. 3, we randomly generate 50 correlation functions with the same photon statistics, using the results $A$ and $Γ$ of the MLE fit to the measured correlation functions. For each of these samples we again perform a MLE fit to yield the new fit results $A'$ and $Γ'$. We use the standard deviation of the resulting distribution of $A'$ as an estimation of the error of $A$. These standard deviations define the error bars along the y axes in Fig. 3.

Theory prediction of $g^{(2)}(0)$ for our experiment. To model the behaviour of the transmitted light as function of atom number shown in Fig. 3, we first estimate the atom number distribution that enters in each data point due to our post-selection on transmission. This distribution is estimated by taking into account the measured probability of preparing an atomic ensemble with certain OD as well as the uncertainty in OD estimation that originates from photon shot noise in the measured transmission. The error bars in the x direction in Fig. 3 indicate the OD spread corresponding to the standard deviation of the atom number distribution. Taking into account this OD spread in the theory34, we calculated the averaged second-order correlation $g^{(2)}(0)$ for a range of discrete ODs. Fitting the resulting dataset to the measurement results shown in Fig. 3 using the coupling strength $β$ as the only fit parameter then yields the theory prediction that is shown as a solid line.

Estimation of $β$. To estimate the coupling strength $β$ of a single atom to the waveguide, we carry out a saturation measurement as described in refs. 24,25. To do so, we send into the nanofibre a probe pulse with a duration of 10 μs with varying power, and record the optical power transmitted through the ensemble. We then fit the absorbed power versus the input power using the generalized Beer–Lambert law with $β$ as a fit parameter. From this, we find $β = 0.0083 ± 0.0003$.

Single-photon output rate. To get an estimate of the photon output rate that can be reached at the point of perfect antibunching, we calculated the power transmission $T$ for the case of a low input photon rate of $n_{\text{in}} ≈ 0.1 × Γ/β$. For $β < 0.1$ this transmission follows approximately $T ≈ β/Γ$. Consequently, the rate of antibunched photons at the output of the ensemble is given by $n_{\text{out}} = T n_{\text{in}} ≈ 0.1 × Γ/β^2 ≈ 0.1 × 1/Γ$. In this regime, after the interaction with the atoms, one obtains a stream of antibunched light with a rate of $0.1/Γ$. For comparison, for a single quantum-emitter-based single-photon source with the same emitter–waveguide coupling strength, one expects a maximum photon rate of $βΓ/2$.

Data availability

The data supporting the findings of this study are available from J.V. (juergen.volz@hu-berlin.de) or A.R. on reasonable request.

Code availability

The code used for modelling the data is available from S.M. on reasonable request.

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Author contributions

K.H., S.M. and A.S.S. made the theory predictions and were responsible for modelling the data. J.H., A.S.P., A.R., S.R., P.S. and J.V. contributed to the design and the setting-up of the experiment. A.S.P. and J.H. performed the experiment. A.S.P. together with S.M. and J.V. was responsible for analysing the data. All authors contributed to the writing of the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

Extended data is available for this paper at https://doi.org/10.1038/s41566-020-0692-z. Correspondence and requests for materials should be addressed to S.M. or A.R. Reprints and permissions information is available at www.nature.com/reprints.
Extended Data Fig. 1 | Second-order correlation functions for different atom numbers. The measured correlation functions are shown in blue and the theoretical predictions (see main text) in orange.
Extended Data Fig. 2 | Experimental sequence. First, a MOT and a optical molasses phase are used to load atoms into the nanofibre-based trap (i). This is followed by a 10 ms ramp of the power of the red-detuned trapping field to increase $\beta$ (ii) and a second molasses phase to re-cool the atoms (iii). To measure the OD of the ensemble, we then sweep the probe laser frequency across the atomic resonance and measure its transmission (iv). The main part of the experimental sequence consists of 350 repetitions of alternating probe and cooling pulses (v). Afterwards, we again measure the OD to check for atom losses (vi), remove the atoms from the trap (vii), and measure transmission through the bare fibre for calibration (viii).