Entanglement with negative Wigner function of almost 3,000 atoms heralded by one photon

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Quantum-mechanically correlated (entangled) states of many particles are of interest in quantum information, quantum computing and quantum metrology. Metrologically useful entangled states of large atomic ensembles have been experimentally realized1–16, but these states display Gaussian spin distribution functions with a non-negative Wigner quasiprobability distribution function. Non-Gaussian entangled states have been produced in small ensembles of ions17–19, and very recently in large atomic ensembles20–22. Here we generate entanglement in a large atomic ensemble via an interaction with a very weak laser pulse; remarkably, the detection of a single photon prepares several thousand atoms in an entangled state. We reconstruct a negative-valued Wigner function—an important hallmark of non-classicality—and verify an entanglement depth (the minimum number of mutually entangled atoms) of 2,910 ± 190 out of 3,100 atoms. Attaining such a negative Wigner function and the mutual entanglement of virtually all atoms is unprecedented for an ensemble containing more than a few particles. Although the achieved purity of the state is slightly below the threshold for entanglement-induced metrological gain, further technical improvement should allow the generation of states that surpass this threshold, and of more complex Schrödinger cat states for quantum metrology and information processing. More generally, our results demonstrate the power of heralded methods for entanglement generation, and illustrate how the information contained in a single photon can drastically alter the quantum state of a large system.

Entanglement is now recognized as a resource for secure communication, quantum information processing, and precision measurements. An important goal is the creation of entangled states of many-particle systems while retaining the ability to characterize the quantum state and validate entanglement. Entanglement can be verified in a variety of ways; one of the strictest criteria is a negative-valued Wigner function23,24, which necessarily implies that the entangled state has a non-Gaussian wavefunction. To date, the metrologically useful spin-squeezed states1–10 have been produced in large ensembles. These states have Gaussian spin distributions and therefore can largely be modelled as systems with a classical source of spin noise, where quantum mechanics enters only to set the amount of Gaussian noise. Non-Gaussian states with a negative Wigner function are however manifestly non-classical, since the Wigner function as a quasiprobability function must remain non-negative in the classical realm. Whereas before this work a negative Wigner function had not been attained for atomic ensembles, in the optical domain, a negative-valued Wigner function has very recently been measured for states with up to 110 microwave photons14. Another entanglement measure is the entanglement depth19, that is, the minimum number of atoms that are demonstrably, but possibly weakly, entangled with one another. This parameter quantifies how widely shared among the particles an entangled state is. For a state of an ensemble characterized by collective measurements, the entanglement depth depends sensitively on the proximity of the state to the ideal symmetric subspace of all particles. The largest entanglement depth verified previously has been 170 out of 2,300

Figure 1 | Scheme for heralded entanglement generation in a large atomic ensemble by single-photon detection. a, Incident vertically polarized light (top left) experiences weak polarization rotation $\phi$ due to atomic quantum noise, and the detection of a horizontally polarized transmitted photon $|\psi\rangle$ heralds an entangled state of collective atomic spin. An optical resonator formed by two mirrors enhances the polarization rotation and the heralding probability. The Bloch spheres show the ideal Wigner distribution functions for the collective spin $S$ upon registering the corresponding single-photon detection events. b, Atoms of $^{87}$Rb in the $5S_{1/2}$ manifold are coupled to the excited $5P_{3/2}$ manifold via linearly polarized light, decomposed into two circular polarization components $|\sigma^{+}\rangle$ (indicated by red arrows) that interact with the atomic ground-state populations. The outgoing polarization state of the light reflects the quantum fluctuations between the $|5S_{1/2}F=1, m=\pm 1\rangle$ magnetic sublevels. See Methods for details.

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atoms for a spin-squeezed state, and very recently 13 out of 41 atoms for a non-Gaussian state.

Here we generate entanglement in a large atomic ensemble by detecting a single photon that has interacted with the ensemble. An incident vertically polarized photon experiences a weak random polarization rotation associated with the quantum noise of the collective atomic spin. The detection of a horizontally polarized emerging photon then heralds a non-Gaussian entangled state of collective atomic spin (Fig. 1) with a negative-valued Wigner function of $-0.36 \pm 0.08$, and an entanglement depth of 90% of our ensemble containing several thousand atoms.

The pertinent atom–light interaction is enhanced by an optical cavity, into which we load $N_c = 3,100 \pm 300$ laser-cooled $^{87}$Rb atoms (Fig. 1a). The atoms are prepared in the $5S_{1/2}, F = 1$ hyperfine manifold, such that each atom $i$ can be associated with a spin $f_i$, and the ensemble with a collective-spin vector $S = \sum f_i$. After polarizing the ensemble ($S_z = S = F = N_c$) by optical pumping, the collective spin state is rotated onto the $x$ axis by means of a radio-frequency $\pi/2$ pulse. This (unentangled) initial state, which is centred about $S_z = 0$ with a variance $(\Delta S_z)^2 = S/2$, is known as a coherent spin state (CSS). In our experiment, the atoms are not uniformly coupled to the optical mode used for state preparation and detection, but the relevant concepts can be generalized to this situation, as discussed in Methods.

Probe light resonant with a cavity mode and detuned from the $^{87}$Rb $D_2$ transition is polarization-analysed upon transmission through the cavity. The vertical polarization state of each photon in the incident laser pulse $|\psi\rangle = (|\sigma^+\rangle + |\sigma^-\rangle)/\sqrt{2}$ can be decomposed into two circular polarization components $|\sigma^\pm\rangle$ that produce opposite differential light shifts between the atomic magnetic sublevels $(m = \pm 1)$. Hence a $|\sigma^\pm\rangle$ photon causes a precession of the collective spin vector $S$ in the $x$–$y$ plane by a small angle $\pm \phi$ (see Methods), and we denote the corresponding slightly displaced CSS by $|\pm \phi\rangle$. Then the combined state of the atom–light system after the passage of one photon can be written as

$$|\psi\rangle \sim |\sigma^+\rangle |\phi\rangle + |\sigma^-\rangle |-\phi\rangle$$

Conversely, atoms in the states $(m = \pm 1)$ cause different phase shifts on the $\sigma^\pm$ photons, resulting in a net rotation of the photon linear polarization if the states $(m = \pm 1)$ are not equally populated. Then the atomic quantum fluctuations between $(m = \pm 1)$ in the CSS randomly rotate the polarization of the input photons $|\psi\rangle$, giving rise to a non-zero probability $\propto \phi^2$ for an incident $|\psi\rangle$ photon to emerge in the polarization $|\phi\rangle = (|\sigma^+\rangle - |\sigma^-\rangle)/\sqrt{2}$, orthogonal to its input polarization. The detection of such a ‘heralding’ photon projects the atomic state onto $|\psi\rangle \propto |\phi\rangle$ or $-|\phi\rangle$, which is not a CSS, but an entangled state of collective spin, namely, the first excited Dicke state $|\Delta_1\rangle$ along $\hat{x}$ (Fig. 1a). In contrast, if the photon is detected in its original polarization $|\psi\rangle$, the atomic state is projected onto $|\psi\rangle \propto |\phi\rangle + |-\phi\rangle$, a state slightly spin squeezed and essentially identical to the input CSS. Thus the entangled atomic state $|\psi\rangle$ is post-selected by the detection of the heralding photon $|\phi\rangle$.

From a different perspective, the entangled state is generated by a single-photon measurement event. The incident photon undergoes Faraday rotation by an angle $\theta$ proportional to the collective spin along the cavity axis, $S_z$, that exhibits quantum fluctuations around $\langle S_z \rangle = 0$. Since detection of the outgoing photon in $|\phi\rangle$ is only possible if $S_z \neq 0$, such detection excludes values of $S_z$ near 0 from the spin distribution, and biases the collective spin towards larger values of $|S_z|$. This creates a ‘hole’ in the atomic distribution near $S_z = 0$, as seen in Fig. 1a.

The mean photon number in the incident laser pulse $k \approx 210$ is chosen such that the probability for one photon to emerge in heralding polarization $|\phi\rangle$ is $p = 0.05 \pm 0.01$. This ensures a very small probability $\propto \phi^2$ for producing a different entangled state $|\psi\rangle$ heralded by two photons, a state which, owing to our photon detection efficiency of $q = 0.3 < 1$, would (mostly) mistake for $|\psi\rangle$. This admixture of $|\psi\rangle$ to the heralded state is suppressed by a factor of $3p(1-q) \approx 0.1$. Further state imperfection arises from false heralding events due to residual polarization impurity of the probe beam (independent of the atoms) of $\sim 3 \times 10^{-5} = 0.1p/k$, adding an admixture of about 10% of the CSS to the heralded state.

In order to reconstruct the collective-spin state generated by the heralding event, we rotate the atomic state after the heralding process by an angle $\beta = 0.15 \pm 0.05 \pi/2$ about the $x$ axis before measuring $S_z$. (Thus $\beta = 0$ corresponds to measuring $S_y$, $\beta = \pi/2$ corresponds to $S_x$, and so on.) The measurement is performed by applying a stronger light pulse in the same polarization-optimized set-up used for heralding. As the Faraday rotation angle $\beta \leq 1$ is proportional to the probability

$$\beta \approx 0.1p/k.$$

Figure 2 | Collective-spin distribution of atomic state heralded by one photon. a–d, Measured photon distributions $g(n_j)$ for no heralding photon detected (blue squares), and for one heralding photon detected (red circles), for rotation angles $\beta$ of 0 (a), $\pi/4$ (b), $\pi/2$ (c) and $3\pi/4$ (d). Inset, logarithmic representations of the same data. In the ideal case, the ratio of the heralded state and the CSS is $g(n_j)^{\text{her}} / g(n_j)^{\text{CSS}} = (S_y)^2 / (S_y)^2 = 3$ for any angle $\beta$, and we measure $g(n_j)^{\text{her}} / g(n_j)^{\text{CSS}} = (2.7 \pm 0.2, 2.2 \pm 0.2, 2.4 \pm 0.2, 2.1 \pm 0.1)$ for $\beta = 0, \pi/4, 3\pi/4$. For each $\beta$, the blue and red data sets represent approximately $1.5 \times 10^4$ and 200 experiments, respectively. The solid blue and the dashed red curves are predictions without any free parameters, calculated from first principles and the separately measured atom number, for the CSS and the perfect first Dicke state, respectively. The solid red line corresponds to the simultaneous fit to all measurement angles $\beta$, that is, the reconstructed density matrix. Error bars, 1 s.d. The planes through the Bloch spheres indicate the measurement direction for the collective spin as specified by the angle $\beta$. e–h, Reconstructed collective spin distributions of the heralded state (red) for rotation angles $\beta$ of 0 (e), $\pi/4$ (f), $\pi/2$ (g) and $3\pi/4$ (h). The spin distributions of the CSS (blue) are for reference. The horizontal axis $S_y$ is expressed in terms of the effective atom number $N' = (2/3)N_c = 2,100$, obtained by weighting each atom with its coupling strength to the standing-wave probe field inside the cavity, such that the experimentally measured spin fluctuation $(\Delta S_y)^2$ of the CSS via its interaction with the probe light satisfies the standard relation $(\Delta S_y)^2 = S/2 = NF/2$ for spin $F$ atoms (see Methods). The shaded area indicates the statistical uncertainty of 1 s.d. The spin distribution in $f$ shows no ‘hole’ in the middle owing to the lower quality of data for this measurement run, $\beta = \pi/4.$
for detecting $|b\rangle$-polarized photons is proportional to $\beta^2$, the measured probability distribution of $|b\rangle$ photon number, $g(n_b)$, reflects the probability distribution of $S_n^b$. Figure 2a–d shows that a single heralding photon substantially changes the spin distribution towards larger values of $|S_{1/2}^\downarrow\rangle$. We further verify that the heralded state remains (nearly) spin polarized with a contrast of $C = 0.99 \pm 0.01 - 0.02$, the same as for the CSS within error bars (Fig. 3a).

From the photon distributions $g(n_b)$ we can reconstruct the density matrix $\rho_{\text{mea}}$ in the Dicke state basis $|n\rangle$ along $\hat{x}$, where $|n\rangle = |0\rangle$ denotes the CSS along $\hat{x}$, $|n\rangle = |1\rangle$ the first Dicke state, $|n\rangle = |2\rangle$ the second Dicke state, and so on. From the density matrix we obtain the Wigner function $W(\theta, \phi)$ on the Bloch sphere (Fig. 3). To accurately determine the Wigner function value on the axis, $W(\theta = \frac{\pi}{2}, \phi = 0) = \sum_n (-1)^n \rho_{nn}$, which depends only on the population terms $\rho_{nn}$ we average the photon distributions $g(n_b)$ over four angles $\beta$ and thereby reduce the fitting parameters to just $\rho_{nn}$, $n \leq 4$. This is equivalent to constructing a rotationally symmetric Wigner function from the angle-averaged marginal distribution $^{17}$. We obtain $\rho_0 = 0.32 \pm 0.03$, $\rho_{11} = 0.66 \pm 0.04$ with negligible higher-order population terms, giving $W(\frac{\pi}{2}, 0) = -0.36 \pm 0.08$, to be compared to $W(\frac{\pi}{2}, 0) = -1$ for the perfect first Dicke state.

We can also fit the density matrix including the coherence terms simultaneously to $g(n_b)$ for all four angles $\beta$, without angle-averaging. Since the photon distributions $g(n_b)$ depend only on $S_2$, they determine only the even terms of the density matrix, that is, $\rho_{nn}$ where $m + n$ is even, and contain no information about the odd terms. If we calculate $W(\frac{\pi}{2}, 0)$ from the density matrix without angle-averaging, we find $W(\frac{\pi}{2}, 0) = -0.27 \pm 0.08$, within error bars consistent with the angle-averaged value. In order to display the Wigner function, we bound the odd terms ($m + n$ odd) by verifying that the heralding process does not displace the state relative to the CSS (see Methods). Therefore we set the odd terms to zero, and display the resulting density matrix and corresponding Wigner function in Fig. 3b–d. The spin distributions $f(S_n)$ obtained from this density matrix are shown in Fig. 2e–h.

In order to quantify the minimum number of mutually entangled atoms, we use a criterion derived in ref. 13 that establishes entanglement depth as a function of the populations $\rho_{00}$ and $\rho_{11}$. From this criterion, generalized to the case of non-uniform coupling to the measurement light field (see Methods), we deduce an average entanglement depth of $N_\text{avg} = 2.910 \pm 190$ out of $N = 3,100$ atoms (Fig. 3e) using the angle-averaged density matrix. Our results represent the first (to our knowledge) experimental verification of the mutual entanglement shared by virtually all atoms in an ensemble that contains more than a few particles.

The above results demonstrate that even with limited resources, that is, weak atom–photon coupling, heralding schemes can be used to boost the effective interaction strength by a large factor, enabling the production of highly entangled states $^{20,23}$. Furthermore, by repeated trials and feedback the entanglement generation can be made quasi-deterministic $^{24,25}$. Our approach is related to other heralded schemes for quantum communication $^{24,27}$ and entangled-state preparation $^{28,30}$, and it would be interesting to generalize the present analysis to infer characteristics of the atomic state from the measured optical signals in those experiments. We note that the same first Dicke state was created in an ensemble of up to 41 atoms with a scheme that uses many heralding photons in a strongly coupled atom–cavity system $^{13}$. In our system, the maximum atom number of $\sim 3,000$ is set by the accuracy of the spin rotation, and could be increased by two orders of magnitude by better magnetic-field control $^{10}$. The state purity $\rho_{11}$ can probably be further improved by reducing the heralding probability, and a value of $\rho_{11} > 0.73$ would be required for the Fisher information $^{14}$ to exceed that of the CSS, and enable metrological gain of up to 3 dB. The detection of two or more photons prepares Schrödinger cat states $^{30}$ of the atomic ensemble with

Figure 3 | Reconstruction of the heralded many-atom entangled state.

(a) Normalized spin component $S_x/S$ measured in a Ramsey sequence, as a function of the phase of the second Ramsey n/2 pulse, for the CSS (blue squares) and the heralded state (red circles). The fit (red line) shows a contrast of $0.99 \pm 0.01 - 0.02$ for the heralded state, within error bars the same as the contrast $0.995 \pm 0.004$ of the CSS. The negligible contrast reduction is expected given that we send only 210 photons into the system at large detuning from atomic resonance. (b) Reconstructed Wigner function $W(\theta, \phi)$ for the heralded state on the Bloch sphere $^{22}$ with a radius given by the effective atom number $N = 2,100$. $\theta$ is the polar angle with respect to $\hat{z}$ and $\phi$ is the azimuthal angle with respect to $\hat{x}$. The first excited Dicke state and the CSS have $W(\frac{\pi}{2}, 0) = -1$ and $W(\frac{\pi}{2}, 0) = 1$, respectively. To provide a reference scale for the size of the negative region, the black dashed line is the contour at which the CSS has a Wigner function value equal to 1, i.e. $e = 0$. (c) Real and imaginary parts, respectively, of the reconstructed density matrix elements, in the Dicke state basis along $\hat{x}$, for the heralded state. (d) Entanglement depth criterion $^{11}$ for the heralded state, plotted in terms of density matrix elements $\rho_{00}$ and $\rho_{11}$. The red shaded region represents the 1 s.d. confidence region for the heralded state. Lines (orange, green and blue, labelled with $k$) represent boundaries for $k$-particle entanglement in terms of atom number $N_k$; a state with $\rho_{11} > 0.73$ would be required for the Fisher information $^{14}$ to exceed that of the CSS, and enable entanglement gain of up to 3 dB. The detection of two or more photons prepares Schrödinger cat states $^{20}$ of the atomic ensemble with
more metrological gain. We expect that heralded methods can generate a variety of nearly pure, complex, strongly entangled states that are not accessible by any other means at the present state of quantum technology.

Online Content Methods, along with any additional Extended Data display items and Source Data, are available in the online version of the paper; references unique to these sections appear only in the online paper.

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METHODS

Photon polarization rotation by atomic spin. Probe laser light red-detuned by $\Delta r/(2\pi) = -200$ MHz from the $^{85}$Rb transition $S^\text{F}{1,2}/C_0/C_1$, $F = 1$ to $S^\text{F}{0,1}/C_0/C_1$, $F = 0$ is sent through an optical cavity containing the atomic ensemble. We first consider the case where all the atoms are coupled with equal strength to the probe light. For detuning $\Delta r$ much larger than the excited state linewidth $\Gamma/(2\pi) = 6.1$ MHz, the excited state manifold can be adiabatically eliminated. The vector component of the a.c. Stark shift is described by the Hamiltonian

$$H_{\text{shift}} = g_\text{s}^2 J_{\text{S}}^z$$

(2)

where $J_{\text{S}}^z = \frac{1}{2} (a^\dagger a + a^\dagger a - a^\dagger a - a^\dagger a)$, with $a$, the annihilation operator for photons with $\sigma^+$ circular polarizations. Here $g_\text{s}$ is the effective single-photon Rabi frequency $\Omega_\text{R}(\gamma)$ to satisfy two conditions: the experimental measured variance equals that of $N$ uniformly coupled atoms, $N_{\text{eff}}/(N_{\text{eff}}^2) = \frac{N}{2} g^2$, and that the total amount of interaction between the atomic ensemble and the probe light is the same, that is, $\langle N_\text{inter} \rangle = N_\text{eff}$. To satisfy these two conditions we define the effective atom number $N = \frac{2}{3} N_\text{eff}$ and the effective cavity cooperativity $\eta = \frac{3}{2} \eta_\text{c}$. This re-scaling allows direct comparison with the well-known expressions for the uniformly coupled CSS.

As in the main text and the rest of Methods, $S_{\text{e}}$ refers to the collective spin of an ensemble containing $N$ effective atoms, and therefore the atomic spin precession phase for each transmitting cavity photon is given by $\phi = \eta \Gamma(4\Delta r/3) = \eta \Gamma(4\Delta r/3)$. Note that this value $\eta = 0.05 < 1$ corresponds to the weak atom–cavity coupling regime. For our parameters, $\phi = 5 \times 10^{-4} \phi_\text{CSS} = 1.5 \times 10^{-5}$ where $\phi_\text{CSS} = 1/(1/25)$ is the angular r.m.s. width of the CSS.

Choice of the heralding photon number. The heralding light must be weak enough that it does not introduce substantial decoherence of the desired atomic state. The fundamental shot noise between the $\sigma^+$ and $\sigma^-$ circular polarization components of the heralding light gives rise to phase broadening of the atomic state, which limits the purity of the heralded entangled state. To measure the phase broadening, heralding light pulses with variable photon number are sent into the cavity, and the variance $\Delta^2_{\text{S}}$ is measured by applying a radio-frequency $\pi/2$ pulse to rotate the atomic state about the $x$ direction before measuring $\Delta^2_{\text{S}}$. Extended Data Fig. 1 shows the measured atomic state variance $\Delta^2_{\text{S}}$ as a function of the photon number in the heralding light, in agreement with the predicted linear dependence. The heralding photon number is thus chosen to be $\leq 210$, with corresponding herald detection probability $p_h = 1.5\%$, to give fairly small phase broadening. Lower heralding photon number results in a purer heralded state, but at the expense of a lower heralding and state generation probability.

Relation between $f(S_{\text{e}})$ and $g(n)$. To measure the atomic state spin distribution, measurement light with the same polarization $\nu$ as the heralding light is sent through the atoms, and the number of photons with the orthogonal polarization $|h|$ is measured. The measurement light contains a large number of input photons $n_{\text{in}} = 1.7 \times 10^4$ to perform destructive measurements with good signal-to-noise ratio. The photon polarization is rotated by a small angle $\hat{\delta} = \delta_\text{S}$, and the probability for each photon to emerge in $|h\rangle$ is $\hat{p}_h$. For a given number of input photons $n_{\text{in}}$, the average number of detected photons with $|h\rangle$ polarization is $n_S = n_{\text{in}} \delta^2_\text{S}$, where $\delta_\text{S}$ is the overall quantum efficiency. Therefore, a spin distribution $f(S_{\text{S}})$ is mapped to a measured photon distribution $g(n_S)$. For a given $S_{\text{S}}$, the detected photons follow a Poisson distribution with mean number $n_S$ and the probability to measure exactly $n$ photons is given by

$$P(n_S) = \exp \left[ -n_S \delta^2_\text{S} \right] \frac{(n_S \delta^2_\text{S})^n}{n!}$$

(5)

For an atomic state with the spin distribution $f(S_{\text{S}})$, the photon distribution $g(n)$ is given by

$$g(n) = \sum_S f(S_{\text{S}}) P(n_S) = \sum_S f(S_{\text{S}}) \exp \left[ -n_S \delta^2_\text{S} \right] \frac{(n_S \delta^2_\text{S})^n}{n!}$$

(6)

In order to measure the spin along a general direction, the atomic spin is rotated by an angle $\beta$ with a radio-frequency pulse before detection. Replacing $S_{\text{S}}$ by $S_{\beta h}$ in equation (6), we write the relation between the spin distribution $f(S_{\beta h})$ and the measured photon distribution $g(n_{\beta h})$ as

$$g(n_{\beta h}) = \sum_S f(S_{\beta h}) P(n_{\beta h} | S_{\beta h}) = \sum_S f(S_{\beta h}) \exp \left[ -n_{\beta h} \delta^2_\text{S} \right] \frac{(n_{\beta h} \delta^2_\text{S})^n}{n!}$$

(7)

Choice of the measurement photon number. The measurement photon number is chosen to optimize the readout quality. Extended Data Fig. 2 illustrates the dependence on readout on the input measurement photon number $n_{\text{in}}$ by showing how the reconstructed distributions $f(S_{\beta h})$ change as $n_{\text{in}}$ is varied (the method of reconstruction is discussed later). When the photon number is small, there is large detection noise due to photon shot noise, reflected as the large error band. With increasing photon number, the photon scattering by atoms into free space increases and the atomic state is more strongly perturbed, therefore the ‘dip’ at $S = 0$ becomes less distinct. To balance these two competing effects, the optimized atomic-state-measurement photon number is set to $1.7 \times 10^4$.

Subtracting background photon counts. Owing to the residual polarization impurity of the measurement light, there are a small number of background photon counts even when there are no atoms. The background counts account for about 4% of the photon signal of the heralded state. We independently measure the background photon distribution and subtract it from the directly measured atomic signal to obtain $g(b_S)$. If we were not to correct for these background counts, we would overestimate the density matrix population $\rho_1$, by 10%.
Reconstruction of the density matrix. Using the measured photon distributions \( g(n_j) \) for all four angles \( \beta = 0, \pi/4, \pi/2, 3\pi/4 \), the density matrix \( \rho \) of the heralded state can be reconstructed.

As the entangled state maintains 0.99 \( ^\pm 0.01 \) contrast, the magnitude of the total spin \( S = N \) and we can express the density matrix in the basis of Dicke states \( |m_n \rangle \) along the \( x \) direction

\[
\rho = \rho_{00}|0 \rangle_0\langle 0| + \rho_{11}|1 \rangle_1\langle 1| + \rho_{01}|0 \rangle_0\langle 1| + \rho_{10}|1 \rangle_1\langle 0| \\
+ \rho_{02}|0 \rangle_0\langle 2| + \rho_{20}|2 \rangle_2\langle 0| + \rho_{12}|1 \rangle_1\langle 2| + \rho_{21}|2 \rangle_2\langle 1| + \ldots
\]

(8)

The spin distribution \( f(|\psi \rangle) \) can be written as a function of atom number \( N \) and the density matrix elements \( \rho_{nm} \), and so on:

\[
f(S, p, \rho | R) = \sum_{N=0}^{\infty} \sum_{m,n} f(S_m | \rho_N) \rho_{nm} \exp[-q_{mn}(S_p)^2] / \sigma_n^2
\]

(11)

Here \( G(m, S_p) = \sum_{N} f(S_p | \rho_N) \rho_{nm} \) is the wavefunction of Dicke state \(|m_n \rangle \) in the representation of spin component \( S_p \) and is given by

\[
G(m, S_p) = \frac{1}{\sqrt{N^m m!}} \left( \frac{1}{2N} \right)^{1/4} e^{-S_p^2/4N} H_m \left( \sqrt{1/N} S_p \right)
\]

(10)

where \( H_m(x) \) is the \( m \)th order Hermite polynomial and \( N \) is the atom number. Using equation (7), we write the theoretically predicted photon distribution \( g_{\eta}(n_j) \) as a function of the density matrix \( \rho \), atom number \( N \) and input photon number \( n_m \)

\[
g_{\eta}(n_j | \rho, N, n_m) = \sum_{S_p} f(S_p | \rho_N) \exp[-q_{mn}(S_p)^2] / \sigma_n^2
\]

(12)

Since the photon distributions \( g(n_j) \) measure \( S_p \), we can obtain the even terms of the density matrix \( \rho_{mn} \), where \( m+n \) is even, and our measurements are not sensitive to the odd terms. Because the odd population probability is \( \rho_{odd} = 0.01 \), the higher-order Dicke-state components are exponentially suppressed. We fit the density matrix up to Dicke state \(|4 \rangle \). The fitted values \( \rho_{12} = 0.03 \pm 0.02 \), \( \rho_{21} = 0.02 \pm 0.01 \), \( \rho_{20} = 0.01 \pm 0.01 \) agree with the theoretical expectation\(^7\) for our system.

From the fitted density matrix \( \rho \), we can obtain the even terms of the density matrix \( \rho_{mn} \) and the atom number \( N \) by fitting the photon distributions of the CSS, which is the only free parameter, to the theoretical distributions \( g_{\eta}(n_j) \) using equation (9) for all four angles \( \beta \). We do this by minimizing the least squares deviation \( D \) weighted by the error \( \sigma_\eta \) of \( g(n_j) \), given by

\[
D = \sum_{\eta} \sum_{n=0}^{\infty} \left( \frac{g_{\eta}(n_j | \rho, N, n_m)}{\sigma_\eta} - g(n_j | \rho) \right)^2
\]

(13)

Performing a measurement with a probe beam polarized at 45° relative to \( |\psi \rangle \), such that the difference between the measured \( |\psi \rangle \) and \( |\psi \rangle \) photon numbers is proportional to \( S_p \). We find a heralding-light-induced shift \( \Delta S_p = -0.2 \pm 1.6 \), consistent with zero, and very small compared to the CSS r.m.s. width \( (\Delta S_p)_{CSS} \approx 30 \). Therefore we set the odd terms of the density matrix to zero in Fig. 3b–d.

Entanglement depth for finite contrast. Entanglement depth is defined as the minimum number of entangled particles in an ensemble. A fully separable pure state can be written as \( |\psi \rangle = |\psi \rangle_1 \otimes \ldots \otimes |\psi \rangle_L \), where \( L \) is the atom number. A pure \( k \) productable state can be written as \( |\psi \rangle = |\psi \rangle_1 \otimes \ldots \otimes |\psi \rangle_L \), where \( k_1, \ldots, k_L \leq k \). If a state cannot be written as a pure \( (k-1) \)-productable state or a mixed state of \((k-1)\)-productable states, then it has entanglement depth at least \( k \).

We slightly generalize the entanglement criterion derived in ref. 13 to take into account the finite contrast \( C \) of the collective atomic spin in our experiment. The derivation in ref. 13 considers the case in the fully symmetric Dicke subspace of \( N \) atoms, and finds that for a \( k \)-productable state the maximum population of the first Dicke state \( \rho_{11} \) as a function of the CSS population \( P_{0} \) is

\[
\max P_{1} = \frac{P_{0}}{N} \max \left[ \sqrt{k} \max \left( \frac{F_{M-1}(a_{1}, \ldots, a_{M-1}) + \sqrt{k} F_{1}(\sqrt{P_{0}/x})}{1} \right) \right]
\]

(14)

Here \( M = [N/k] \), \( k = N - k(M-1) \), and \( F_{M-1}(a_{1}, \ldots, a_{M-1}) = \sum_{i=1}^{k} \sqrt{1 - a_{i}^{2}} \). Equation (14) is generally not a concave function of \( P_{0} \). In order to obtain the upper bound for mixed states, denote the concave hull of the right side of equation (14) as \( B(P_{0}, k, N) \). We define \( B(P_{0}, k, N) = B(P_{0}, k, N) / N \). Note that when \( N \leq k_{0} \), \( B(P_{0}, k, N) = B(P_{0}, k, N) \).

The heralded state we produce does not necessarily retain perfect contrast, so the state can be a mixture of different total spin states \( S = N - 1, \ldots, N(1 - \epsilon) \), with \( \epsilon = 0.01 \). The contrast loss is mainly caused by the decoherence between \( F = 1 \) magnetic sublevels, and the free space scattering of the heralding light by the atoms. We decompose the density matrix \( \rho \) into the total spin basis

\[
\rho = \sum_{\eta=0}^{\infty} w_{\eta} \rho_{\eta} \rho_{\eta-1}
\]

(15)

Here \( \rho_{\eta-1} \) is the density matrix in the subspace of total spin \( S = N - \eta \), \( w_{\eta} \) is the weight for each \( \rho_{\eta-1} \), and \( \sum_{\eta=0}^{\infty} w_{\eta} = 1 \). For each \( \rho_{\eta-1} \)

\[
B(P_{0}, k, N - i) = B(P_{0}, N - k, N - i) / (N - i) \leq B(P_{0}, N - k, N) / (N - i)
\]

(16)

Here \( B(P_{0}, N - k, N) \) is the probability for the state to be found in the ground state in the subspace of total spin \( N - i \).

Measurements of the spin distributions do not allow us to determine the total spin of the system at single-atom resolution. We define populations of the CSS and the first Dicke state by

\[
P_{0} = \sum_{\eta=0}^{\infty} w_{\eta} \rho_{\eta-N-1}
\]

(17)

\[
P_{1} = \sum_{\eta=0}^{\infty} w_{\eta} \rho_{\eta-N-1}
\]

(18)

The upper bound of \( P_{1} \) is given by

\[
\max P_{1} \leq \sum_{\eta=0}^{\infty} w_{\eta} \max P_{\eta-N-1} \leq \sum_{\eta=0}^{\infty} w_{\eta} B(P_{0}, N - k, N - i) / (N - i)
\]

(19)

Using equation (16) and the fact that \( B(P_{0}, k, N) \) is a concave function of \( P_{0} \) we have

\[
\max P_{1} \leq \sum_{\eta=0}^{\infty} w_{\eta} B(P_{0}, N - k, N - i) / (N - i)
\]

(20)

Here \( C \) is the contrast of the collective spin. Comparing to ref. 13, the result is modified by a factor \( 1/C \). In our experiment, \( C = 0.99 \pm 0.01 \), so the effects of finite contrast on entanglement depth are minimal.
generalize the entanglement criterion to our experimental conditions and prove that the sample-averaged fractional entanglement depth for the ensemble containing 3,100 actual non-uniformly coupled atoms is the same as that of 2,100 uniformly coupled effective atoms. Consider an ensemble of \( N_a \) actual atoms where each atom \( j \) has spin component \( f_{jx} \) and cooperativity \( \eta_j \). The effective total spin of the ensemble is \( S_e \), and the effective cooperativity is \( \eta \), so that

\[
S_e \eta = \sum_{j=1}^{N_a} f_{jx} \times \eta_j \tag{21}
\]

As mentioned in the main text, the ideal heralded state \( |\psi_i \rangle \) (the first Dicke state of non-uniformly coupled atoms) is the destructive interference of two slightly displaced CSSs \( \pm |\psi \rangle \) and can be written as

\[
|\psi_i \rangle \propto |\psi \rangle - |\bar{\psi} \rangle
\]

\[
= \left[ e^{i \frac{\delta}{4} / (4k)} - e^{-i \frac{\delta}{4} / (4k)} \right] |\psi_0 \rangle
\]

\[
= e^{i \delta / (4k)} \sum_i \eta_i |j \rangle |0 \rangle \otimes |1 \rangle_\chi
\]

where \( |\psi_0 \rangle \) is the initial CSS along \( \chi \). By expanding the exponent to first order and using \( f_{0} = (f_{+x} - f_{-x})/2(2) \), we get

\[
|\psi_1 \rangle = \left( \sum_{j=0}^{N_a} \eta_j \right)^{-1/2} \sum_{j=1}^{N_a} \eta_j \left[ |0 \rangle_j \otimes |0 \rangle_\chi + |1 \rangle_j \otimes |1 \rangle_\chi \right]
\]

(23)

where \( |0 \rangle_j \) and \( |1 \rangle_j \) are the single-particle spin eigenstates along \( \chi \) of the atom \( j \). For a fully separable state \( |\psi \rangle = \prod_{j=1}^{N_a} (\xi_j |0 \rangle_j + \beta_j |1 \rangle_j) \ldots \) the population \( P_1 = (|\psi| \langle \psi |)^2 \) is given by

\[
P_1 = \left( \sum_{j=1}^{N_a} \eta_j^2 \right)^{-1/2} \sum_{j=1}^{N_a} \eta_j \beta_j \xi_j
\]

(24)

The expression for \( P_1 \) is similar to that in ref. 13 and differs by the additional weight factor \( \eta_j \). When the real atom number \( N_a \gg 1 \), the upper bound of \( P_2 \) for the fully separable state \( |\phi \rangle \), \( B(P_0, N_a) \), as a function of the population \( P_0 = (|\phi| \langle \phi |)^2 \), is the same as ref. 13, and independent of \( N_a \).

Next consider a state which can be factorized into two subsets \( |\psi \rangle = |\phi_1 \rangle \otimes |\phi_2 \rangle \) where \( k_1 + k_2 = N_a \). Each \( |\phi_i \rangle \) can be expanded as

\[
|\phi_i \rangle = a_i |\phi_{k_1} \rangle + b_i |\phi_{k_2} \rangle + \ldots
\]

(25)

where \( |\phi_{k_1} \rangle \) is the CSS containing \( k_1 \) atoms, and \( |\phi_{k_2} \rangle \) is given by equation (23) with \( N_a \) replaced by \( k_2 \). The populations \( P_0 = (|\phi| \langle \phi |)^2 \) and \( P_1 = (|\phi| \langle \phi |)^2 \) are given by

\[
P_0 = |a_1|^2 |a_2|^2,
\]

\[
P_1 = \left( \sum_{j=1}^{N_a} \eta_j^2 \right)^{-1} a_2 b_1 \left[ \sum_{j=k_1+1}^{N_a} \eta_j^2 + a_1 b_2 \left( \sum_{j=k_1+1}^{N_a} \eta_j^2 \right) \right]
\]

(26)

The expression for \( P_1 \) recovers that of ref. 13 when \( \eta_j = 1 \). When \( k_1, k_2 \) and \( N_a \) are large, we take the ensemble averages \( \sum_{j=k_1+1}^{N_a} \eta_j^2 = k_2 (\eta_j^2) \) and \( \sum_{j=k_1+1}^{N_a} \eta_j^4 = N_a (\eta_j^4) \). Therefore the bound of \( P_1 \) in equation (26), \( B(P_0, k_2) = \max (k_1, k_2), N_a \), is the same as \( B(P_0, k_2, N_a) \) for uniformly coupled atoms when \( k_2 / N_a = k/2N \). This proves that the average fractional entanglement depth for the ensemble containing 3,100 actual non-uniformly coupled atoms is the same as that of 2,100 uniformly coupled effective atoms, thus in our system a minimum of 1,970 out of 2,100 effective atoms or 2,910 out of 3,100 real atoms are mutually entangled.

It might seem as if the addition of \( N_a \gg N \) weakly coupled atoms (coupling strength \( \eta_n \)) to the system would increase the entanglement depth without having physical consequences as long as \( N_a \eta_n^2 \ll N \eta^2 \). However in this case the uncertainty \( \Delta N \) on the entanglement depth also increases, given by

\[
\frac{\Delta N}{N_a} = \frac{\Delta N}{N_a} \eta_n^2 \gg \frac{\Delta N}{N}
\]

so as to be consistent with the entanglement depth before adding the weakly coupled atoms. Atoms that do not change the observed spin distribution have no effect on the entanglement depth.

Sample size. No statistical methods were used to predetermine sample size in the above.

31. Tanji-Suzuki, H. et al. Interaction between atomic ensembles and optical resonators: classical description. Adv. At. Mol. Opt. Phys. 60, 201–237 (2011).
Extended Data Figure 1 | The measured atomic state spin variance, $\Delta S_y^2$, as a function of the heralding light photon number and corresponding probability $pq$ of detecting one photon. The solid red line is the prediction for $\Delta S_y^2$ broadened by the photon shot noise of the heralding light. The dashed black line shows the CSS variance for 2,030 $F = 1$ effective atoms used in this measurement. Error bars, 1 s.d.
Extended Data Figure 2 | Dependence of the reconstructed distribution of collective spin $S_z$ on the measurement photon number. This dependence is illustrated by reconstructed spin distributions for photon numbers $0.5 \times 10^4$ (a), $1.1 \times 10^4$ (b), $1.7 \times 10^4$ (c), $2.7 \times 10^4$ (d) and $3.6 \times 10^4$ (e). Blue lines correspond to the CSS and red lines correspond to the heralded states. The shaded area indicates an uncertainty of 1 s.d.
Extended Data Table 1 | Resonator parameters

| Parameter                  | $\lambda$=780nm | $\lambda$=852nm |
|----------------------------|-----------------|-----------------|
| Mirror separation          | $L$             | 26.62(1) mm     |
| Mirror curvature radius    | $R$             | 25.04(2) mm     |
| Free spectral range        | $\omega_{FSR}/(2\pi)$ | 5632.0(2) MHz   |
| Transverse mode spacing    | $\omega_{\lambda}/(2\pi)$ | 226.3(3) MHz    |
| Linewidth                  | $\kappa_{\lambda}/(2\pi)$ | 1.01(3) MHz     | 135(2) kHz |
| Finesse                    | $F_{\lambda}$   | $5.6(2) \times 10^3$ | $4.2(1) \times 10^4$ |
| Mode waist                 | $w_{\lambda}$   | 56.9(4) $\mu$m  | 59.5(5) $\mu$m |
| Antinode cooperativity     | $\eta_{0,\lambda}$ | 0.203(7)        | 1.65(4)     |

The mode waists are calculated at the position of the atoms. Outside this table, all resonator values refer to the probe wavelength $\lambda = 780$ nm.
Erratum: Entanglement with negative Wigner function of almost 3,000 atoms heralded by one photon
Robert McConnell, Hao Zhang, Jiazhong Hu, Senka Cuk & Vladan Vuletić

In Fig. 2a of this Letter, the solid blue line in the linear plot was inadvertently removed during the production process; this figure has now been corrected in the online versions of the manuscript.