

## Supplementary Materials for

### **First observation of the quantized exciton-polariton field and effect of interactions on a single polariton**

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## Supplementary Materials

### section S1. Interaction Hamiltonian

The interacting character of the polaritons is inherited from their excitonic component. The most general Hamiltonian describing interacting polaritons is given by [25]  $H = H_0 + H_{\text{shift}} + H_{\text{scat}}$ , with (we take  $\hbar = 1$  along the text)

$$H_0 = \sum_{\mathbf{k}, \sigma=\uparrow, \downarrow} \omega_{a, \mathbf{k}} a_{\sigma, \mathbf{k}}^\dagger a_{\sigma, \mathbf{k}} + \omega_{b, \mathbf{k}} b_{\sigma, \mathbf{k}}^\dagger b_{\sigma, \mathbf{k}} + \mathbf{g}_{\mathbf{k}} (a_{\sigma, \mathbf{k}}^\dagger b_{\sigma, \mathbf{k}} + b_{\sigma, \mathbf{k}}^\dagger a_{\sigma, \mathbf{k}}) \quad (\text{S1a})$$

$$H_{\text{shift}} = \sum_{\mathbf{k}, \sigma=\uparrow, \downarrow} V_{\mathbf{k}, \mathbf{k}, 0}^{(1)} b_{\sigma, \mathbf{k}}^\dagger b_{\sigma, \mathbf{k}}^\dagger b_{\sigma, \mathbf{k}} b_{\sigma, \mathbf{k}} + V_{\mathbf{k}, \mathbf{k}, 0}^{(2)} b_{\sigma, \mathbf{k}}^\dagger b_{\sigma, \mathbf{k}}^\dagger b_{-\sigma, \mathbf{k}} b_{-\sigma, \mathbf{k}} \quad (\text{S1b})$$

$$H_{\text{scat}} = \sum_{\substack{\mathbf{k}, \mathbf{k}' \neq \mathbf{k} \\ \sigma=\uparrow, \downarrow}} V_{\mathbf{k}, \mathbf{k}', 0}^{(1)} b_{\sigma, \mathbf{k}}^\dagger b_{\sigma, \mathbf{k}}^\dagger b_{\sigma, \mathbf{k}} b_{\sigma, \mathbf{k}'} + V_{\mathbf{k}, \mathbf{k}', 0}^{(2)} b_{\sigma, \mathbf{k}}^\dagger b_{-\sigma, \mathbf{k}'}^\dagger b_{\sigma, \mathbf{k}} b_{-\sigma, \mathbf{k}'} +$$

$$+ \frac{1}{4} \sum_{\substack{\mathbf{k}, \mathbf{k}' \neq \mathbf{k} \\ \sigma=\uparrow, \downarrow}} \exp(i\Delta_{\mathbf{k}, \mathbf{k}', \delta\mathbf{k}} t) (V_{\mathbf{k}, \mathbf{k}', \delta\mathbf{k}}^{(1)} b_{\sigma, \mathbf{k}+\delta\mathbf{k}}^\dagger b_{\sigma, \mathbf{k}'-\delta\mathbf{k}}^\dagger b_{\sigma, \mathbf{k}} b_{\sigma, \mathbf{k}'} + 2V_{\mathbf{k}, \mathbf{k}', \delta\mathbf{k}}^{(2)} b_{\sigma, \mathbf{k}+\delta\mathbf{k}}^\dagger b_{-\sigma, \mathbf{k}'-\delta\mathbf{k}}^\dagger b_{\sigma, \mathbf{k}} b_{-\sigma, \mathbf{k}'} + \text{h.c.}) \quad (\text{S1c})$$

where  $a_{\sigma, \mathbf{k}}$  ( $b_{\sigma, \mathbf{k}}$ ) is the annihilation operator of the photon (exciton) with polarization  $\sigma$ , momentum  $\mathbf{k}$ , energy  $\omega_{a, \mathbf{k}}$  ( $\omega_{b, \mathbf{k}}$ ) and  $\Delta_{\mathbf{k}, \mathbf{k}', \delta\mathbf{k}} = (E(\mathbf{k}) + E(\mathbf{k}') - E(\mathbf{k} + \delta\mathbf{k}) - E(\mathbf{k}' - \delta\mathbf{k}))/\hbar$ . Here  $H_0$  describes the linear coupling between photons and excitons, which occurs when the two particles have the same polarization and the same momentum.  $H_{\text{shift}}$  describes the interactions between excitons with the same momentum but possibly different polarizations, and  $H_{\text{scat}}$  describes the scattering between excitons with different momenta. The matrix elements  $V_{\mathbf{k}, \mathbf{k}', \delta\mathbf{k}}^{(1)}$  and  $V_{\mathbf{k}, \mathbf{k}', \delta\mathbf{k}}^{(2)}$  describe the scattering of two polaritons in the triplet and the singlet configurations, respectively. The experimental evidence allows us to reduce the number of terms that play a role in the dynamics of an ideal system, namely

- i) The lower polariton branch is completely empty, except at the wavevector  $\mathbf{k} = \mathbf{k}_0$  where a laser creates a polariton condensate. Here we assume that the condensate is in a coherent state which allows us to do a mean field approximation replacing the operators of the lower polariton branch for complex numbers.

- ii) The upper polariton branch is empty, except for the single photon that enters at  $\mathbf{k} = \mathbf{0}$ . This means that the nonlinear terms of the Hamiltonian for the upper polariton branch always evaluate to zero.

The latter condition implies that Eq. (S1b) does not contribute to the dynamics, and that Eq. (S1c) reduces to

$$H_{\text{scat}} = 3V^{(1)}(b_{\uparrow,0}^\dagger b_{\uparrow,0} b_{\uparrow,\mathbf{k}_0}^\dagger b_{\uparrow,\mathbf{k}_0} + b_{\downarrow,0}^\dagger b_{\downarrow,0} b_{\downarrow,\mathbf{k}_0}^\dagger b_{\downarrow,\mathbf{k}_0}) + 2V^{(2)}(b_{\downarrow,0}^\dagger b_{\downarrow,0} b_{\uparrow,\mathbf{k}_0}^\dagger b_{\uparrow,\mathbf{k}_0} + b_{\uparrow,0}^\dagger b_{\uparrow,0} b_{\downarrow,\mathbf{k}_0}^\dagger b_{\downarrow,\mathbf{k}_0}) + V^{(2)}(b_{\uparrow,0}^\dagger b_{\downarrow,0} + b_{\downarrow,0}^\dagger b_{\uparrow,0})(b_{\uparrow,\mathbf{k}_0}^\dagger b_{\downarrow,\mathbf{k}_0} + b_{\downarrow,\mathbf{k}_0}^\dagger b_{\uparrow,\mathbf{k}_0}) \quad (\text{S2})$$

where we have assumed that the interaction energies do not depend on the wavevectors. The lower and upper polaritons are related to the photon and exciton through the transformation,

$$a_{\sigma,\mathbf{k}} = \cos \theta_{\mathbf{k}} q_{\sigma,\mathbf{k}} + \sin \theta_{\mathbf{k}} p_{\sigma,\mathbf{k}} \quad \text{and} \quad b_{\sigma,\mathbf{k}} = \sin \theta_{\mathbf{k}} q_{\sigma,\mathbf{k}} - \cos \theta_{\mathbf{k}} p_{\sigma,\mathbf{k}} \quad (\text{S3})$$

where  $p_{\sigma,\mathbf{k}}$  and  $q_{\sigma,\mathbf{k}}$  are the annihilation operators of the lower and upper polaritons, respectively; and  $\sigma$  is the polarization of the particles. Since the dispersion relation of the photon and of the exciton do not have the same curvature, their contribution to the lower and upper polariton is different at each  $\mathbf{k}$ . This variation is taken into account through the Hopfield coefficient

$$\sin \theta_{\mathbf{k}} = \sqrt{\frac{\xi_{\mathbf{k}}^2}{1 + \xi_{\mathbf{k}}^2}} \quad \text{with} \quad \xi_{\mathbf{k}} = \frac{(\omega_{b,\mathbf{k}} - \omega_{a,\mathbf{k}}) + \sqrt{4g_{\mathbf{k}}^2 + (\omega_{b,\mathbf{k}} - \omega_{a,\mathbf{k}})^2}}{2g_{\mathbf{k}}} \quad (\text{S4})$$

In the lower and upper polariton basis, the Hamiltonian in Eq. (S1a) becomes

$$H_0 = \sum_{\mathbf{k}, \sigma=\uparrow, \downarrow} q_{\sigma,\mathbf{k}}^\dagger q_{\sigma,\mathbf{k}} (\omega_{b,\mathbf{k}} \sin^2 \theta_{\mathbf{k}} + \omega_{a,\mathbf{k}} \cos^2 \theta_{\mathbf{k}} + g_{\mathbf{k}} \cos \theta_{\mathbf{k}} \sin \theta_{\mathbf{k}}) + p_{\sigma,\mathbf{k}}^\dagger p_{\sigma,\mathbf{k}} (\omega_{b,\mathbf{k}} \cos^2 \theta_{\mathbf{k}} + \omega_{a,\mathbf{k}} \sin^2 \theta_{\mathbf{k}} - g_{\mathbf{k}} \cos \theta_{\mathbf{k}} \sin \theta_{\mathbf{k}}) \quad (\text{S5})$$

whereas Eq. (S2) becomes

$$H_{\text{scat}} = 3V^{(1)}(q_{\uparrow,0}^\dagger q_{\uparrow,0} p_{\uparrow,\mathbf{k}_0}^\dagger p_{\uparrow,\mathbf{k}_0} + q_{\downarrow,0}^\dagger q_{\downarrow,0} p_{\downarrow,\mathbf{k}_0}^\dagger p_{\downarrow,\mathbf{k}_0}) \cos^2 \theta_{\mathbf{k}_0} \sin^2 \theta_0 + 2V^{(2)}(q_{\downarrow,0}^\dagger q_{\downarrow,0} p_{\uparrow,\mathbf{k}_0}^\dagger p_{\uparrow,\mathbf{k}_0} + q_{\uparrow,0}^\dagger q_{\uparrow,0} p_{\downarrow,\mathbf{k}_0}^\dagger p_{\downarrow,\mathbf{k}_0}) \cos^2 \theta_{\mathbf{k}_0} \sin^2 \theta_0 + V^{(2)}(q_{\uparrow,0}^\dagger q_{\downarrow,0} + q_{\downarrow,0}^\dagger q_{\uparrow,0})(p_{\uparrow,\mathbf{k}_0}^\dagger p_{\downarrow,\mathbf{k}_0} + p_{\downarrow,\mathbf{k}_0}^\dagger p_{\uparrow,\mathbf{k}_0}) \cos^2 \theta_{\mathbf{k}_0} \sin^2 \theta_0 \quad (\text{S6})$$

where we have taken into account that at  $\mathbf{k} = \mathbf{k}_0$  only the lower polariton branch is populated, whereas at  $\mathbf{k} = \mathbf{0}$  only the upper polariton branch is populated. Resorting again to the fact that the condensate in the lower polariton branch is in a coherent state, the operators for the lower polariton at any given wavevector can be replaced by the squared root of the population at such wavevector, namely  $p_{\sigma,\mathbf{k}} = \sqrt{n_\sigma} \delta_{\mathbf{k},\mathbf{k}_0}$ , where  $n_\sigma$  is the mean number of particles with polarization  $\sigma$  in the lower polariton branch at  $\mathbf{k} = \mathbf{k}_0$ , and  $\delta$  is the Kronecker delta. At last, we include a spin-orbit coupling term that enters into the Hamiltonian as an effective magnetic field that couples polaritons with the same momenta but opposite spins:  $H_{so} = g_{so}(q_{\uparrow,0}^\dagger q_{\downarrow,0} + q_{\downarrow,0}^\dagger q_{\uparrow,0})$  (8). The magnitude of the coupling is given by the imbalance of polaritons with spin  $+1$  and with spin  $-1$  and the polariton interaction strength, namely  $g_{so} = 2(V^{(1)} - V^{(2)})(n_\uparrow - n_\downarrow) \cos^2 \theta_{\mathbf{k}_0} \sin^2 \theta_0$ . After applying the mean field approximation, the resulting Hamiltonian has a simple form

$$H = \omega_\uparrow q_\uparrow^\dagger q_\uparrow + \omega_\downarrow q_\downarrow^\dagger q_\downarrow + g_{\uparrow\downarrow} (q_\uparrow^\dagger q_\downarrow + q_\downarrow^\dagger q_\uparrow) \quad (\text{S7})$$

with

$$\begin{aligned} \omega_\uparrow &= \cos^2 \theta_{\mathbf{k}_0} \sin^2 \theta_0 (3n_\uparrow V^{(1)} + 2n_\downarrow V^{(2)}) + \omega_0 \\ \omega_\downarrow &= \cos^2 \theta_{\mathbf{k}_0} \sin^2 \theta_0 (3n_\downarrow V^{(1)} + 2n_\uparrow V^{(2)}) + \omega_0 \\ g_{\uparrow\downarrow} &= [2\sqrt{n_\uparrow n_\downarrow} V^{(2)} + 2(V^{(1)} - V^{(2)})(n_\uparrow - n_\downarrow)] \cos^2 \theta_{\mathbf{k}_0} \sin^2 \theta_0 \\ \omega_0 &= \sin^2 \theta_0 \omega_{b,0} + \cos^2 \theta_0 \omega_{a,0} + g_0 \cos \theta_0 \sin \theta_0 + \omega_0 \end{aligned} \quad (\text{S8})$$

In the sample used for the experiment, the photon-exciton detuning was 2.3 at  $\mathbf{k} = \mathbf{0}$ , zero at  $\mathbf{k} = \mathbf{k}_0$  and the Rabi coupling (half Rabi splitting) was 1.6 at  $\mathbf{k} = \mathbf{0}$ . Therefore the Hopfield coefficients are

$$\cos^2 \theta_{\mathbf{k}_0} = \frac{1}{2} \quad \text{and} \quad \sin^2 \theta_0 \approx 0.79. \quad (\text{S9})$$

## section S2. Dynamics of the entangled photons

The nonlinear crystal emits pair of photons entangled in their polarization degree of freedom, described by the wavefunction

$$|\psi_0\rangle = \frac{1}{\sqrt{2}}(|\mathbf{H}, \mathbf{V}\rangle + |\mathbf{V}, \mathbf{H}\rangle) \equiv \frac{1}{\sqrt{2}}(c_{\mathbf{H}}^\dagger q_{\mathbf{V}}^\dagger + c_{\mathbf{V}}^\dagger q_{\mathbf{H}}^\dagger)|0\rangle \quad (\text{S10})$$

where  $c_\phi^\dagger$  is the creation operator of the single photon with polarization  $\phi$  that goes directly to the detector, whereas  $q_\phi^\dagger$  is the creation operator of the photon with polarization  $\phi$  that impinges onto the upper polariton branch at  $\mathbf{k} = \mathbf{0}$ . The latter, when inside the cavity, evolves according to Schrödinger equation with the Hamiltonian in Eq. (S7) for the duration  $\Delta t$  of the mean polariton lifetime, producing as the output wavefunction  $|\psi(t)\rangle = \exp(-iH\Delta t)|\psi_0\rangle$ . Note, however, that the parameters of the Hamiltonian depend on the mean number of particles in the lower polariton branch, cf. Eqs. (S8). Such a population is build by a laser. The poissonian fluctuations in the intensity of the laser induces a fluctuation in the number of particles in the condensate. We go beyond a mean-field description to take into account these fluctuations by sampling randomly the state of the condensate when the upper single-polariton is created, and evolving its wavefunction accordingly. Averages are then computed from the set of wavefunctions with the fluctuating Hamiltonian.

Before one of the entangled photons is send to the cavity, the state of the lower and upper polariton branches can be written as

$$|\psi_0\rangle = |n_\uparrow, n_\downarrow\rangle \otimes \frac{i}{\sqrt{2}}(|\uparrow, \uparrow\rangle - |\downarrow, \downarrow\rangle), \quad (\text{S11})$$

where  $n_\uparrow$  ( $n_\downarrow$ ) is the macroscopic population of the lower branch with spin  $\sigma = \uparrow$  ( $=\downarrow$ ) and  $(|\uparrow, \uparrow\rangle - |\downarrow, \downarrow\rangle)/\sqrt{2}$  is the state in Eq. (S10) written in the basis of circular polarization. The state after one of the single photons has passed through the cavity is obtained simply by propagating the initial state with the Hamiltonian in Eq. (S7), from which we obtain

$$|\psi(t)\rangle = e^{-iHt/\hbar} |\psi_0\rangle = |n_\uparrow, n_\downarrow\rangle \otimes [c_{\uparrow, \uparrow}(t)|\uparrow, \uparrow\rangle + c_{\downarrow, \downarrow}(t)|\downarrow, \downarrow\rangle + c_{\uparrow, \downarrow}(t)(|\uparrow, \downarrow\rangle - |\downarrow, \uparrow\rangle)] \quad (\text{S12})$$

where the coefficients are related to the detuning and the coupling strength through the following relations,

$$c_{\uparrow,\uparrow}(t) = \frac{1}{\sqrt{2}R} \left[ \Delta \sin\left(\frac{Rt}{2}\right) + iR \cos\left(\frac{Rt}{2}\right) \right], \quad c_{\downarrow,\downarrow}(t) = c_{\uparrow,\uparrow}^*(t), \quad \text{and} \quad c_{\uparrow,\downarrow}(t) = \sqrt{2} \frac{g_{\uparrow,\downarrow}}{R} \sin\left(\frac{Rt}{2}\right) \quad (\text{S13})$$

where we have used the shorthand notation  $\Delta = \omega_{\uparrow} - \omega_{\downarrow}$  and  $R = \sqrt{4g_{\uparrow,\downarrow}^2 + \Delta^2}$ . The final state of the system is obtained by averaging over many realizations, and because of the fluctuations the state becomes mixed and the correlations are washed out. In fact, when  $Rt/2$  is uniformly distributed around an interval  $\delta_{\theta}$  the purity is given by

$$\text{Tr}(\rho^2) = \frac{1}{2} + \frac{1 - \cos \delta_{\theta}}{\delta_{\theta}^2} \geq \frac{1}{2} \quad (\text{S14})$$

The effect of the condensate in the lower branch on the state of the single photon that impinges onto the upper branch, can be obtained by propagating the initial state of the single photon, i.e.,  $|\psi(0)\rangle = i(|\uparrow\rangle - |\downarrow\rangle)/\sqrt{2}$ , with the effective Hamiltonian in Eq. (S7)

$$|\psi(t)\rangle = e^{-iHt/\hbar} |\psi(0)\rangle = \frac{1}{\sqrt{2}} \left[ i \cos\left(\frac{Rt}{2}\right) - \frac{2g_{\uparrow,\downarrow} + \Delta}{R} \sin\left(\frac{Rt}{2}\right) \right] |\uparrow\rangle - \frac{1}{\sqrt{2}} \left[ i \cos\left(\frac{Rt}{2}\right) - \frac{2g_{\uparrow,\downarrow} - \Delta}{R} \sin\left(\frac{Rt}{2}\right) \right] |\downarrow\rangle \quad (\text{S15})$$

and the probability to measure the single photon in the state  $|\varphi\rangle = (\alpha|\uparrow\rangle + \beta e^{i\theta}|\downarrow\rangle)/N$  with  $N = \sqrt{\alpha^2 + \beta^2}$  is given by

$$P_{\varphi} = |\langle \varphi | \psi(t) \rangle|^2 = \frac{1}{2NR^2} \left[ R^2 (N^2 - 2\alpha\beta \cos \theta) \cos^2\left(\frac{Rt}{2}\right) + 4\alpha\beta R \Delta \sin \theta \cos\left(\frac{Rt}{2}\right) \sin\left(\frac{Rt}{2}\right) + (R^2 \diamond^2 + 4(\alpha^2 - \beta^2)g_{\uparrow,\downarrow}\Delta - 2\alpha\beta(R^2 - 2\Delta^2) \cos \theta) \sin^2\left(\frac{Rt}{2}\right) \right] \quad (\text{S16})$$

where  $R$  and  $\Delta$  are as in Eq. (S13).

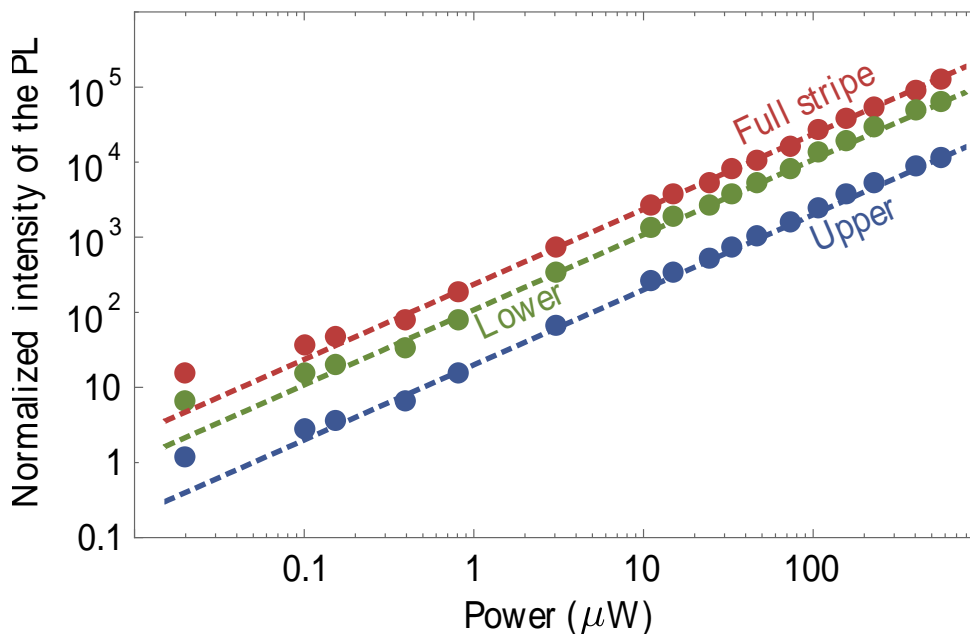
### section S3. Effect of noise on the measurement

The behaviour shown in Figure 5 was obtained by performing a *hyper-complete tomography* (41) of the state of the pair of entangled photons. Since such a tomography consists of counting the coincidences of the photons in a combination of three bases of polarization (logical, diagonal and circular) (35), as discussed in the *Sample & Setup* section in the main text, the noise in the form of accidental coincidences from any source not related to the entangled photons can be corrected on

average by subtracting background coincidences measured in absence of the quantum source. There is another form of noise, however, that cannot be corrected in this way in presence of the condensate. The increasing power on the  $x$ -axis of Fig. 5 corresponds to more populated polariton condensates in the microcavity, that collectively affects the single polariton sent by the quantum source. The more dense is the condensate, the higher is its effect on the concurrence. The interpretation for this observation is, as explained in the text, that more polaritons in the condensate lead to stronger shifts to the energy of the single polariton due to polariton-polariton interactions, spoiling concurrence through fluctuations of the number of condensed polaritons. Another factor that would affect concurrence in absence of interactions with the single polariton, is if the condensate, at  $k = k_0$  on the lower branch, would scatter particles to the mode excited by the quantum source, i.e., to the upper branch at  $k = 0$ . Such a “noise” would populate the mode excited by the entangled-photon with polaritons that are indistinguishable from the single-polariton created by the quantum source. This noise cannot be corrected by simple subtraction of a background, as it is not “unrelated” to the signal, being indistinguishable from it. For instance, would an entangled photon and such a scattered polariton be present simultaneously in the cavity, we should expect coalescence effects such as Bose stimulation, which is in fact, additional physics rather than noise. The respective rates at which the mode is populated by scattered polaritons, on the one hand, and by the quantum source, on the other hand, suggest that the effect should however be very small. But since the effect on the concurrence could be large, we have to account for it in the theoretical modelling. We now discuss how doing so allows us to rule out that our observation is dominated by scattered polaritons from the condensate and that the observation is explained instead by direct interactions of the single upper polariton at  $k = 0$  with the rest of the lower-polariton condensate at  $k = k_0$ . To quantify the amount of polaritons that are scattered to the wavevector of the single polariton, we measured the luminescence from the cavity. The wavevector selection was made by spatially filtering the emission with a pinhole. Then, the energy resolution was obtained By passing the light through a monochromator. In fig. S1, we show the intensity of the emission at the wavevector from which we collected the photons to count the coincidences with the single photon. There, we show in red the emission without filtering in energy (as the APD is not sensible to the energy of the photons that it collects), in green the contribution only from the lower polariton branch and in blue the contribution only from the upper polariton branch. These polaritons are in fact populating the state to which the single photon arrives, and thus they are reducing the amount of correlated signal.

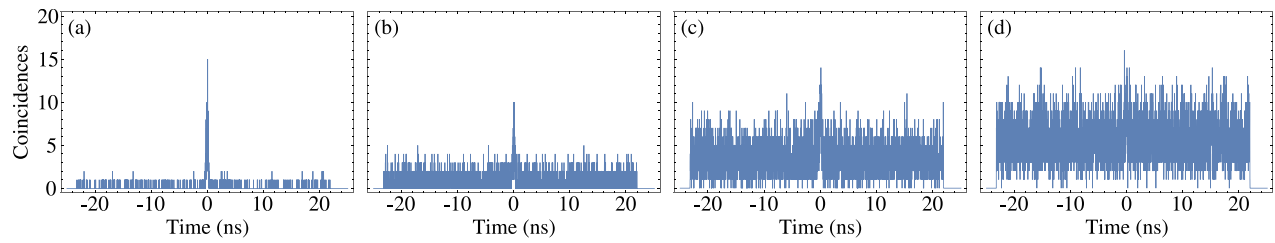
Figure S2 shows, for the various pumping powers create a condensate in the cavity, the coincidences between photons emitted by the cavity with vertical polarization and photons going directly to the APD with horizontal polarization. This basis corresponds to the states in which the photons are prepared in a superposition, and thus provides a maximum visibility for the signal. In panel (a), corresponding to the lowest power used in the experiment, the accidental coincidences (constant background) can be clearly distinguished from the correlated coincidences (peak at zero delay). However, such a distinction becomes less clear as the intensity of the driving laser increases, producing more background, as we show in the progression from Panel (b) to Panel (d). The observation of the luminescence in fig. S1 indicates the need to upgrade the idealized description introduced previously. The presence of the scattered polaritons in the wavevector at which the single photon arrives can be described in the theoretical model as a continuous incoherent pumping of polaritons to such a wavevector. The number of corresponding polaritons can be estimated from fig. S1: for a laser intensity of 20, the intensity of the luminescence from the lower branch is about 2000 times larger than the intensity due to a single polariton, which comes with a repetition rate of 500 000 photon pairs per second. Since the cavity has a lifetime of 3, the average number of photons at the wavevector where the single polariton impinges is thus 0.003. The description of the system is therefore upgraded to a master equation, in which the rate

of the incoherent pumping is chosen so that the steady state population of the cavity is equal to the average population estimated from the luminescence. To take into account only the coincidental detection, we use the Quantum Monte Carlo method (47) and compute the quantum trajectories of the experiment. In them, the Hamiltonian propagation is still done by Eq. (S7), while the decay and incoherent driving are taken into account as quantum jumps. After many such trajectories, we reconstruct the state of the entangled photons by performing the tomography, in the same way that we reconstruct the state from the actual experiment. In Fig. 5 of the main text, we show the dependence of the concurrence on the intensity of the laser driving in the lower branch, taking into account the noise due to the scattered polaritons (green, dashed line) and both the noise and the interactions (blue, solid line). It is clear that if the state to which the entangled polariton arrives is largely populated, the tomography of the pair of photons becomes spoiled and the concurrence drops. However, the noise due to the scattered polaritons from the condensate is far from enough to account for our observations. This assures that interactions are the dominating effect.



**fig. S1. Luminescence of the sample at the wave vector at which the single photon impinges normalized to the intensity of the single polariton.** The colour lines corresponds to the full stripe (red), namely without filtering in energy the emission of the sample and including the scattered photons too, the light emitted from the lower branch (green) and from the upper branch (blue). The measured values are shown by the dots, while the dotted lines represent the linear fit of the data. Note that when the concurrence of the entangled photons starts to fall (at about  $1 \mu\text{W}$ ) the intensity of the luminescence from the lower branch is about 200 times as larger as the single photons.





**fig. S2. Counting photon coincidences as a function of pumping power.** The photon that passes through the cavity and becomes a polariton is detected with vertical polarization. The other one, that goes directly from the nonlinear crystal to the APD is measured in with horizontal polarization. The laser sustaining the polariton condensate impinges onto the cavity has a power of (a) 50 nW, (b) 5  $\mu$ W, (c) 16  $\mu$ W and (d) 26  $\mu$ W. In all the panels the background noise follows a Poissonian distribution. As the power of the driving laser increases, the amount of coincidences due to the entangled single polariton are not simply added on top of the background noise, but instead they remain constant. This follows from the “noise” produced by scattered polaritons from the condensate to the mode excited by the quantum source, being indistinguishable with the signal, and requiring being included in the model rather than subtracted from the data.