Self-homodyne-enabled generation of indistinguishable photons: supplementary material

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METHODS

Sample fabrication: The MBE-grown structure consists of an ≈ 900 nm thick $\text{Al}_{0.8}\text{Ga}_{0.2}\text{As}$ sacrificial layer followed by a 145 nm thick GaAs layer that contains a single layer of InAs QDs. Our growth conditions result in a typical QD density of $(60 - 80) \mu\text{m}^{-2}$. Using 100 keV e-beam lithography with ZEP resist, followed by reactive ion etching and HF removal of the sacrificial layer, we define the photonic crystal cavity. The photonic crystal lattice constant was $a = 246$ nm and the hole radius $r \approx 60$ nm. The cavity fabricated is a linear three-hole defect (L3) cavity with a measured quality factor of $Q = 17700$. To improve the cavity quality factor, holes adjacent to the cavity were shifted [1, 2].

Optical spectroscopy: All optical measurements were performed with a liquid helium flow cryostat at temperatures in the range 10 – 40 K. For excitation and detection a microscope objective with a numeric aperture of $NA = 0.75$ was used. Cross-polarised measurements were performed using a polarising beam splitter. To further enhance the extinction ratio, additional thin film linear polarisers were placed in the excitation/detection pathways and a single mode fibre was used to spatially filter the detection signal. Furthermore, two waveplates were placed between the beamsplitter and microscope objective: a half-wave plate to rotate the polarisation relative to the cavity and a quarter-wave plate to correct for birefringence of the optics and sample itself. We estimate our overall efficiency to be $\sim 0.125\%$, including setup losses and detector efficiency. Excluding the detector efficiency this corresponds to a collection efficiency into the detection single mode fiber of $\sim 0.5\%$

Correlation measurements: Second-order autocorrelation

measurements were performed using a Hanbury Brown and Twiss (HBT) setup consisting of one fibre beamsplitter and two single-photon avalanche diodes. The detected photons were correlated with a PicoHarp300 time counting module. Two-photon interference measurements were performed with the same detectors and electronics as the second-order correlation measurements but with a Mach-Zehnder interferometer replacing the single beamsplitter, as discussed in the main text. For the unfiltered two-photon interference measurements the integration time was 30 minutes and the coincidence rate (in the whole histogram) was 2336/s. For the frequency filtered two-photon interference measurements the integration time was 8.6 hours and the coincidence rate (in the whole histogram) was 22/s.

DETAILS ON THE SIMULATIONS

The quantum-optical simulations were performed using density matrix master equations with the Quantum Optics Toolbox in Python (QuTiP)[3], where the standard[4] Jaynes-Cummings model was used as a starting point. The effects of phonons were incorporated through the addition of incoherent decay channels with rates that were previously extracted[5].

To simulate the first order spectra of our system under excitation with a single pulse, we compute the one-sided spectrum

$$S(\omega) = \text{Re} \left[\iint_{\mathbb{R}^2} d\tau d\tau' e^{-i\omega\tau} \langle A^\dagger(t+\tau) A(t) \rangle \right] \quad (1)$$

of the free-field mode operator $A(t)$. Input-output theory can relate the internal cavity mode operator $a(t)$ to the external field operator by the radiative cavity field decay rate $\kappa/2$. Hence, for a JC system in the solid state where the QD radiative decay

rate γ plays an insignificant role compared to κ [5], spectral decomposition of the cavity mode operator yields the spectrum of the detected light. Therefore, we can compute an unnormalised version of this spectrum with $A(t) \rightarrow a(t)$ in equation (1). We can also compute an unnormalised version of the incoherent spectrum with $\langle A^\dagger(t+\tau)A(t) \rangle \rightarrow \langle A^\dagger(t+\tau)A(t) \rangle - \langle A^\dagger(t+\tau) \rangle \langle A(t) \rangle$ in equation (1). To arrive at the version measured by a spectrometer of finite bandwidth, we convolve $S(\omega)$ with the spectrometer's response function. To simulate self-homodyne suppression (SHS), we replace $A(t) \rightarrow a(t) + \alpha(t)$ in equation (1). Physically, $\alpha(t)$ is a slightly phase- and amplitude-shifted version of the incident laser pulse (originating from the continuum-mode scattering)[6].

In order to simulate the normalised measured degree of second-order coherence, $g^{(2)}[0] = \frac{G^{(2)}[0]}{N^2}$ with $N = \int_{\mathbb{R}} dt \langle A^\dagger(t)A(t) \rangle$, we calculate

$$g^{(2)}[0] = \frac{\int \int_{\mathbb{R}^2} dt d\tau \langle \mathcal{T}_- [A^\dagger(t)A^\dagger(t+\tau)] \mathcal{T}_+ [A(t+\tau)A(t)] \rangle}{\left(\int_{\mathbb{R}} dt \langle A^\dagger(t)A(t) \rangle \right)^2} \quad (2)$$

under excitation by a single pulse[7]. The operators $\mathcal{T}_\pm$ indicate the time ordering required of a physical measurement[8]. We can likewise replace $A(t) \rightarrow a(t)$ in equation (2) and also model SHS with the replacement of $A(t) \rightarrow a(t) + \alpha(t)$ in equation (2). Despite the simplicity of equation (1), adding spectral filtering to equation (2) is analytically and numerically quite challenging. The spectral decomposition of this equation requires a fourth order integral that is often intractable even numerically. Fortunately, the newly discovered sensor formalism[8] allows for efficient calculation of the spectrally filtered version of the measured degree of second-order coherence. Here, we coherently attach a pair of two-level sensors to the system Hamiltonian with the addition of the sensor Hamiltonian to the Jaynes-Cummings Hamiltonian:

$$H = H_{JC} + \sum_{i=1}^2 \left[\omega_s \zeta_i^\dagger \zeta_i + \epsilon \left(a \zeta_i^\dagger + a^\dagger \zeta_i \right) \right] \quad (3)$$

where ω_s is the sensor frequency, ζ the sensor annihilation operator, and ϵ the sensor coherent coupling strength. The sensor coupling is chosen small enough so that its backaction on the system is negligible, i.e. $\frac{\epsilon^2}{\Gamma^2} \ll \gamma_f$ where γ_f is the fastest transition rate in the un-sensed system. Additionally, the sensor decay terms of rate Γ are added to the total Liouvillian. Here, in order to simulate SHS, we replace

$$a \zeta_i^\dagger + a^\dagger \zeta_i \rightarrow (a + \langle \alpha(t) \rangle) \cdot \zeta_i^\dagger + (a^\dagger + \langle \alpha^*(t) \rangle) \cdot \zeta_i \quad (4)$$

in equation (3).

To arrive at the physically measured and spectrally filtered second-order coherence functions, the total degree of second-order coherence is computed between the two sensors:

$$g^{(2)}[0] = \frac{\int \int_{\mathbb{R}^2} dt d\tau \langle \mathcal{T}_- [\zeta_1^\dagger(t) \zeta_2^\dagger(t+\tau)] \mathcal{T}_+ [\zeta_1(t+\tau) \zeta_2(t)] \rangle}{\left(\int_{\mathbb{R}} dt \langle \zeta_1^\dagger(t) \zeta_1(t) \rangle \right)^2} \quad (5)$$

As the sensors are degenerate in every manner, the ordering of their operation is arbitrary. In our model, the sensors are used as filters while the detector is assumed to be sufficiently broadband to integrate the correlations over our entire experimental domain. This approximation is accurate as the detector has a timing resolution of greater than 200 ps compared with the system decay time of approximately 65 ps. Additionally, we integrate our data over ≈ 400 ps bins.

Fig. S1. Incoherent spectra. Power dependent spectra for resonantly exciting UP1 with 16 ps short pulses at a QD-cavity detuning of $\Delta = 4.5 g$.

SIMULATED POWER-DEPENDENT SPECTRA

We began our simulations by computing the incoherent power spectrum to find the π pulse driving power. As all experiments were performed with a π pulse driving, we wanted to ensure an accurate match with simulations. Since the Rabi oscillations are clearly present in the incoherent spectrum without having to optimise SHS, this served as a simple way to verify the π pulse power. The result of such a simulation is presented in Fig. S1 for a resonant excitation of UP1 with 16 ps short pulses at a QD-cavity detuning of $\Delta = 4.5g$. For this detuning, the forward (UP1 to LP1) and reverse (LP1 to UP1) phonon-assisted transfer rates are $2\pi \cdot 2.4$ GHz and $2\pi \cdot 2.2$ GHz respectively.

Note that after optimizing SHS, a nearly identical simulation of the measured spectrum is observed (see main text). For both UP1 and LP1 Rabi oscillations are observed and the pulse area for a π pulse is close to $\sqrt{\frac{\pi}{2}} \frac{\Delta}{2g\tau_p}$ where τ_p is the width parameter of the Gaussian pulse. This analytic result comes from truncating the detuned Jaynes-Cummings level structure and calculating the Raman transition frequency between the lowest-lying levels[6]. From there, the π pulse driving strength is given approximately by integrating the pulse area - the exact drive area is difficult to analytically calculate due to the nonlinear damping of the system.

HONG-OU-MANDEL INTERFERENCE

We first review interference in a Hong-Ou-Mandel (HOM) interferometer. In such an interferometer, two identically independent sources are interfered on two detectors by the action of a single beamsplitter. This setup is shown schematically in Fig. S2. Here, we consider two strongly coupled systems each driven with the same pulse $\Omega(t)$. Their outputs, the Heisenberg free-field operators $a(t)$ and $b(t)$, are then fed into the beamsplitter which mixes the two according to the unitary transformation

$$\begin{bmatrix} c(t) \\ d(t) \end{bmatrix} = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 & -1 \\ 1 & 1 \end{bmatrix} \begin{bmatrix} a(t) \\ b(t) \end{bmatrix} \quad (6)$$

Fig. S2. Schematic of the Hong-Ou-Mandel interferometer. In this interferometer, two driven strongly coupled systems are interfered on two detectors by a beamsplitter. A digital recorder then correlates the detection times and computes $E[n_c[t_1]n_d[t_2]]$. Here, $\Omega(t)$ indicates the coherent driving field, $a(t)$ and $b(t)$ indicate the continuous mode free-field operators at the outputs of the driven systems, and $c(t)$ and $d(t)$ indicate the free-field operators at the inputs to the detectors.

Fig. S3. Schematic of the Mach-Zehnder interferometer. In this interferometer, a single driven strongly coupled system is doubly excited at an interval of $T = 1.9$ ns, to be interfered with a time-delayed copy of itself. Importantly, the re-excitation only occurs after all excited population from the first pulse has decayed, so that the operator $a'(t)$ is identically independent between excitations. The vacuum mode operator at the input to the first beamsplitter has been omitted in anticipation that our detectors only measure a normally ordered moment of their input fields. A digital recorder then again correlates the detection times and computes $E[n_c'[t_1]n_d'[t_2]]$. Here, $\Omega'(t) = \Omega(t) + \Omega(t - T)$ indicates the coherent driving field, $a'(t) = a(t) + a(t - T)$ indicates the continuous mode free-field operator at the output of the driven system, and $c'(t)$ and $d'(t)$ indicate the free-field operators at the inputs to the detectors.

The detection events are then correlated electronically to arrive at a temporal coincidence histogram $E[n_c[t_1]n_d[t_2]]$. In terms of the instantaneous correlations aforementioned, we wish to calculate $E[n_c[t_1]n_d[t_2]] = G_{cd}^{(2)}[0]$. In order to perform this calculation, we first decompose the underlying correlations of $G_{cd}^{(2)}(t_1, t_2)$ into more manageable components based on $a(t)$ and $b(t)$ that will allow us to average over all quickly varying phase terms. The phase terms are difficult to observe experimentally since they depend on femtosecond phase locking and are, regardless, not required for the observation of two-photon interference. It has been shown that after performing this procedure[9] one arrives at

$$G_{cd}^{(2)}(t_1, t_2) = \frac{1}{2}G_a^{(2)}(t_1, t_2) + \frac{1}{2}\left[G_a^{(1)}(t_1, t_1) \cdot G_a^{(1)}(t_2, t_2) - \left|G_a^{(1)}(t_1, t_2)\right|^2\right] \quad (7)$$

where the correlations between $c(t)$ and $d(t)$ have terms only coming from a single source (the indices could be trivially switched as $a(t)$ and $b(t)$ are identically independent). Experimentally due to unknown detection efficiencies, our only true intensity reference is in proportion to the source correlation N_a^2 . Combining this information with the discussion on detector bandwidth above, we are justified to compute only the integrated and normalised correlation

$$g_{\text{HOM}}^{(2)}[0] = \frac{1}{2}g_a^{(2)}[0] + \frac{1}{2}\left[1 - \left|g_a^{(1)}[0]\right|^2\right] \quad (8)$$

referenced in the main text (equation [1]). Hence the observable HOM interference for indistinguishable sources simply consists of an instantaneous measurement that depends on the total first- and second-order coherence of the source. Whether this interference is truly just two-photon interference therefore depends on the nature of the source output. Additionally, we note that in many real systems the first-order coherence extracted here is wildly different than the first-order coherence extracted from a Michelson interferometer due to the long-time averaging action of the Michelson interferometer.

MACH-ZEHNDER INTERFERENCE

Experimentally, producing a single source of indistinguishable photons is often quite challenging (let alone two), however we would still like to characterise the source's instantaneous degree of first order coherence. By doing so, we hope to quantify how it would behave in a HOM experiment. It was realised that some aspect of two-photon interference can be observed for a single source at the input to a Mach-Zehnder (MZ) interferometer[10]. In order to observe this interference, the system must be doubly excited at a time interval that matches the temporal delay between the two paths of the MZ interferometer (shown schematically in Fig. S3). Additionally, this interval must be long enough that the emission resulting from the two excitations is identically independent.

For perfect single photon input the MZ output correlation is in fact two-photon interference and thus equivalent to HOM interference. However, in many cases the literature has incorrectly

compared the two when the source has some finite probability of emitting non-single photons. Thus, we now outline the derivation of the correct measured correlation, as discussed in the main text (equation [2]). Consider the schematic in Fig. S3: A single strongly coupled system is driven with the coherent driving field $\Omega'(t) = \Omega(t) + \Omega(t - T)$ and has field-mode operator output $a'(t) = a(t) + a(t - T)$. The first beamsplitter simply splits $a'(t)$ in two. Here we have dropped the vacuum operator in anticipation that our detectors only measure a normally ordered moment of their input fields. Next, one path is delayed by the excitation interval $T = 1.9$ ns. The second beamsplitter then mixes $\frac{1}{\sqrt{2}}a'(t)$ with the time delayed version of itself and the detection apparatus computes $E[n_{c'}[t_1]n_{d'}[t_2]] = G_{c'd'}^{(2)}[0]$ like before.

The time delay is absolutely critical: the purpose is that for time delays much less than T , $\frac{1}{\sqrt{2}}a'(t)$ can interfere with itself, and therefore reproduce a correlation similar to the HOM cross-correlation. Although computation of $g_{c'd'}^{(2)}[0]$ in terms of source correlations certainly appears daunting, the independence of $a'(t)$ for times greater than T dramatically simplifies the calculation of $g_{c'd'}^{(2)}[0]$. This expansion is further simplified by consideration of the correlations centred around time delays that are integer multiples of T . It is fairly trivial to show that the majority of terms for delays larger than T are zero, with only a few additional phase-dependent terms arising. We note that although one might expect the correlations to be phase locked by the MZ interferometer, our integration times of many hours quickly destroy the phase interference. Since these terms once again play no role in two-photon interference, we made no effort to further stabilise our setup to observe them.

Consider the correlations about zero delay: $a'(t)$ and its time delayed version are statistically independent so equation (7) holds but with the source correlations of $a'(t)$ instead of $a(t)$. Because the source is doubly excited the ratio of the correlations in terms of $a(t)$ is altered, which can easily be seen by applying the above rules. Now, we arrive at the measured degree of correlation

$$G_{c'd'}^{(2)}[0] = G_a^{(2)}[0] + \frac{1}{2} \left[N_a^2 - |G_a^{(1)}[0]|^2 \right] \quad (9)$$

choosing instead to normalise to the maximum value of this expression we arrive at the equation discussed in the main text

$$g_{MZ}^{(2)}[0] = \frac{2}{3}g_a^{(2)}[0] + \frac{1}{3} \left[1 - |g_a^{(1)}[0]|^2 \right] \quad (10)$$

Next, we discuss the influence of non-equal splitting ratios at the beamsplitters to arrive at the correlations comprising each of the five observed peaks. As presented in the main text, we built our Mach-Zehnder (MZ) interferometer from fiber-coupled beam splitters and polarisation maintaining fibers. While this realization of a MZ interferometer is very stable, the commercially available fiber-coupled beam splitters have an imperfect transmittivity (T) to reflectivity (R) ratio that deviates from 50 : 50. Therefore, we need to derive the amplitudes of the five peak pattern observed in the correlation measurements with the individual transmittivities and reflectivities of the first beamsplitter (T_1, R_1) and second beamsplitter (T_2, R_2) included. Both of our beamsplitters have a $T : R$ ratio of 0.56 : 0.44. The transmitted component at the first beamsplitter is sent to the longer arm of the interferometer. Importantly, since this delay is realised

Fig. S4. Two photon interference. Raw data of the frequency filtered MZ measurements.

Fig. S5. Influence of pulse length. Simulation of $g^{(2)}[0]$ obtained from a two-level system under resonant excitation with π pulses of variable length.

with an additional fiber there is an additional coupling loss resulting in an effective splitting ratio of the first beamsplitter of $T_1 = R_1 = 0.44 = B$. Then, the amplitudes of the five peaks in the correlation measurements with SPCM2 as start and SPCM1 as stop are given by:

$$A_1 = AB^2 R_2^2 \quad (11)$$

$$A_2 = AB^2 \left(2R_2 T_2 + 2R_2^2 g_a^{(2)}[0] \right) \quad (12)$$

$$A_3 = AB^2 \left(4R_2 T_2 g_a^{(2)}[0] + \left(R_2^2 + T_2^2 - 2|g_a^{(1)}[0]|^2 R_2 T_2 \right) \right) \quad (13)$$

$$A_4 = AB^2 \left(2R_2 T_2 + 2T_2^2 g_a^{(2)}[0] \right) \quad (14)$$

$$A_5 = AB^2 T_2^2 \quad (15)$$

where A is the absolute amplitude. These equations can now be used to fit the measured data. We determine AB^2 from fitting a number of peaks away from zero time delay using $g_a^{(2)}[0] = 1$ and $|g_a^{(1)}[0]|^2 = 0$, followed by fitting the five peaks around zero time delay. Note that when binning the data also an extremely weak dark-count noise was subtracted.

A binned version of the frequency filtered two-photon interference measurements was presented in Fig. 2d of the main text. For completeness, the raw data of this measurement is presented in Fig. S4.

LOWER LIMIT OF $G^{(2)}[0]$

Finally, we discuss the non-zero value of $g^{(2)}[0]$. For strongly coupled QD-cavity systems $g^{(2)}[0]$ strongly depends on the pulse length, which has to be chosen as a compromise between re-excitation during the presence of the pulse and spectral overlap of the laser with higher climbs up the JC ladder [5]. However, when using SHS and frequency filtering the situation simplifies and becomes identical to that of a two-level system, where the only limitation is re-excitation which is determined by the pulse length. To visualize this dependence, we present in Fig. S5 a simulation of $g^{(2)}[0]$ obtained from a two-level system under resonant excitation with π pulses as a function of the pulse length. Clearly, $g^{(2)}[0]$ increases with pulse length and very small values are only reached for pulses that are much shorter than the state lifetime.

REFERENCES

1. Y. Akahane, T. Asano, B.-S. Song, and S. Noda, "Fine-tuned high-q photonic-crystal nanocavity," *Opt. Express* **13**, 1202–1214 (2005).
2. M. Minkov and V. Savona, "Automated optimization of photonic crystal slab cavities," *Scientific Reports* **4** (2014).
3. J. Johansson, P. Nation, and F. Nori, "Qutip 2: A python framework for the dynamics of open quantum systems," *Computer Physics Communications* **184**, 1234 – 1240 (2013).
4. P. Alsing, D.-S. Guo, and H. J. Carmichael, "Dynamic stark effect for the jaynes-cummings system," *Phys. Rev. A* **45**, 5135–5143 (1992).
5. K. Müller, K. A. Fischer, A. Rundquist, C. Dory, K. G. Lagoudakis, T. Sarmiento, Y. A. Kelaita, V. Borish, and J. Vučković, "Ultrafast polariton-phonon dynamics of strongly coupled quantum dot-nanocavity systems," *Phys. Rev. X* **5**, 031006 (2015).
6. K. A. Fischer, K. Müller, A. Rundquist, T. Sarmiento, A. Y. Piggott, Y. Kelaita, C. Dory, K. G. Lagoudakis, and J. Vučković, "Self-homodyne measurement of a dynamic mollow triplet in the solid state," *Nat Photon* **10**, 163–166 (2016).
7. K. Müller, A. Rundquist, K. A. Fischer, T. Sarmiento, K. G. Lagoudakis, Y. A. Kelaita, C. Sánchez Muñoz, E. del Valle, F. P. Laussy, and J. Vučković, "Coherent generation of non-classical light on chip via detuned photon blockade," *Phys. Rev. Lett.* **114**, 233601 (2015).
8. E. del Valle, A. Gonzalez-Tudela, F. P. Laussy, C. Tejedor, and M. J. Hartmann, "Theory of frequency-filtered and time-resolved n -photon correlations," *Phys. Rev. Lett.* **109**, 183601 (2012).
9. M. J. Woolley, C. Lang, C. Eichler, A. Wallraff, and A. Blais, "Signatures of Hong–Ou–Mandel interference at microwave frequencies," *New J. Phys.* **15**, 105025 (2013).
10. C. Santori, D. Fattal, J. Vučković, G. S. Solomon, and Y. Yamamoto, "Indistinguishable photons from a single-photon device," *Nature* **419**, 594–597 (2002).