

Indistinguishable Photons from a Two-Photon Cascade

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Decay of a four-level diamond scheme via a cascade is a potential source of entangled photon pairs. A solid-state implementation is the biexciton cascade in a semiconductor quantum dot. While high entanglement fidelities have been demonstrated, the two photons, XX and X, are temporally correlated, typically resulting in poor photon coherence. Here, we demonstrate a high two-photon interference visibility (a measure of the photon coherence) for both XX ($90 \pm 2\%$) and X ($80 \pm 6\%$) photons. This is achieved by Purcell enhancing the biexciton transition in a low-noise device. We find that the interference visibility follows the well-known quantum optics result upon tuning the XX:X lifetime ratio over 2 orders of magnitude. In addition, we show that the XX purity, 98%, is limited not by quantum optics but by the phonons in the semiconductor matrix via cavity feeding.

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Entangled photons are a key resource in quantum communication [1]. For basic quantum key distribution following the E91 protocol, a two-photon entangled state is sufficient [2]. More elaborate schemes involve entanglement swapping for which the photons in separate pairs must be indistinguishable [3]. Parametric down-conversion using a nonlinear crystal can be used to create such pairs but such sources are probabilistic [4,5]. A deterministic source of indistinguishable entangled photon pairs, particularly one operating at a high repetition rate, would represent a radical improvement. An option here is to use a semiconductor quantum dot (QD). A QD has a number of attractive features: the optical dipole moments are large such that operation at gigahertz rates is feasible [6]; the interaction with phonons is weak such that the photons are coherent; noise can be suppressed in state-of-the-art heterostructures [7]; and photonic engineering can be used to create high efficiencies [8–13]. However, while creating indistinguishable single photons with a QD has been achieved, an

outstanding challenge is to create highly indistinguishable, entangled photon pairs.

A charge-neutral QD can be doubly excited to populate the QD with a biexciton, a spinless complex of two electrons and two holes. The biexciton decays to the exciton ($|XX\rangle \rightarrow |X\rangle$) emitting the XX photon; the exciton decays to the ground state ($|X\rangle \rightarrow |0\rangle$), emitting the X photon, Fig. 1(a). The two orthogonally polarized excitons result in two (orthogonally polarized) decay paths for the $|XX\rangle$, opening up a scheme to create an entangled photon pair [14,15]. On eliminating the splitting between the two exciton states (the “fine structure”) [16], which-path information is erased and the two photons are entangled in the polarization basis [17]. This scheme has been widely implemented and in the best case yields very high entanglement fidelities, up to 98.7% [17–19]. Alternatively, one decay path can be used to create an entangled photon pair in the time-bin basis [20]: the system should be prepared in a metastable state, perhaps a “dark” exciton [21], with an optical transition to the biexciton. The biexciton is then created using a $(\pi/2)$ pulse (“early” time bin) followed by a π pulse (“late” time bin). Work is underway to implement this alternative scheme [22]. With respect to photon coherence, both schemes suffer from the same drawback. Inevitably, the XX photon is detected before the X photon resulting in an unwanted temporal correlation and a loss of photon coherence (as quantified via the indistinguishability metric, the two-photon interference visibility V) [23].

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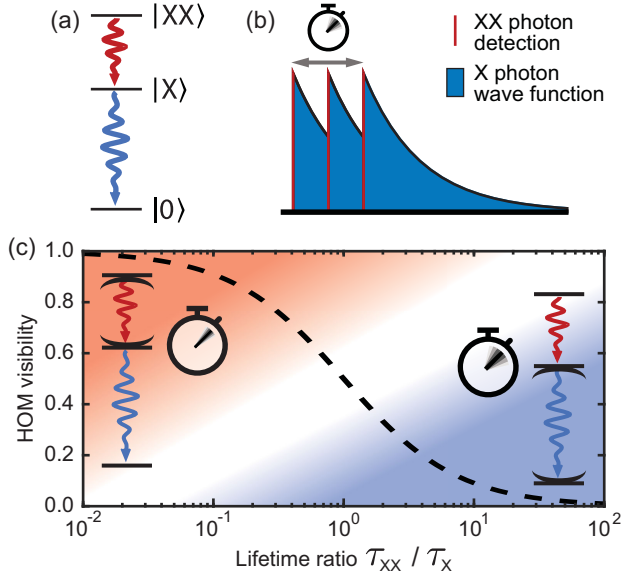


FIG. 1. (a) Schematic of the biexciton cascade. The biexciton decays to the exciton ($|XX\rangle \rightarrow |X\rangle$) emitting photon XX; the exciton decays to the ground state ($|X\rangle \rightarrow |0\rangle$) emitting photon X. Only one decay pathway is shown for simplicity. (b) The XX and X photons are correlated in time limiting the indistinguishability of successive XX photons, likewise X photons. The sketch shows a simplified picture: detection of the XX results in timing jitter in the X photon. (c) Theoretical prediction of the Hong-Ou-Mandel (HOM) interference visibility as a function of lifetime ratio, Eq. (1). τ_{XX} (τ_X) is the radiative lifetime of the $|XX\rangle$ ($|X\rangle$). Reducing τ_{XX} by Purcell enhancing $|XX\rangle \rightarrow |X\rangle$ reduces the timing jitter and improves the coherence of both XX and X photons (red shaded area). Conversely, reducing τ_X has the opposite effect (blue shaded area).

In simple terms, timing jitter reduces the coherence of both XX and X photons, Fig. 1(b).

Quantum optics (Wigner-Weisskopf theory with no biexciton or exciton dephasing) predicts that the coherence metric V for both photons is given by [20,23,24]

$$V = \frac{1}{1 + \tau_{XX}/\tau_X}, \quad (1)$$

where τ_{XX} is the radiative lifetime of the biexciton and τ_X the radiative lifetime of the exciton. In the standard case, $\tau_{XX} \approx 0.65\tau_X$ [25], $V \approx 0.6$, a modest value. Equation (1), points to a powerful mitigation strategy: a cavity can be used to manipulate τ_{XX} and τ_X via the Purcell effect [26], Fig. 1(c). Once $\tau_{XX} \ll \tau_X$, V is close to unity. On QDs, Purcell factors of up to 40 have been achieved [27] such that V is potentially as high as 98.8%. Equation (1) has not been probed experimentally over a wide range of lifetime ratio τ_{XX}/τ_X . It is an open question to what extent Eq. (1) applies to the biexciton cascade given that the theory neglects all the complexities of the semiconductor (biexciton or exciton phonon scattering, charge and spin noise). Previous attempts at controlling V relied either on

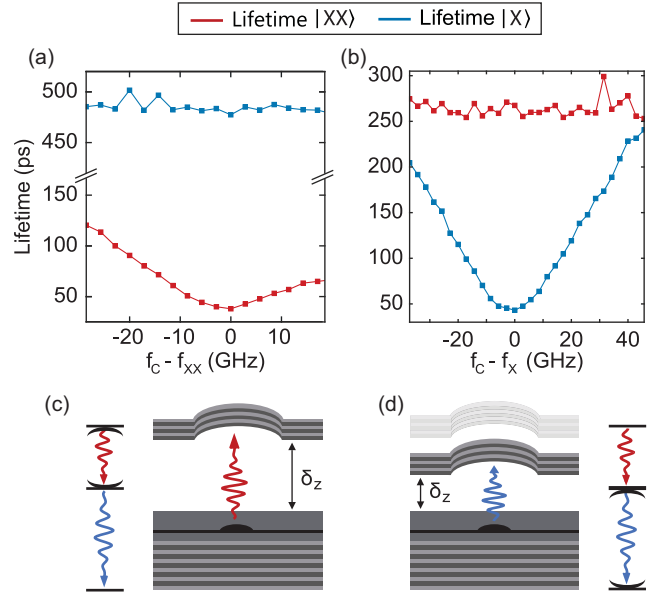


FIG. 2. Transition lifetimes detecting the X and XX photons upon scanning the cavity resonance over (a) $|XX\rangle \rightarrow |X\rangle$ and (b) $|X\rangle \rightarrow |0\rangle$. f_c is the cavity resonance frequency, f_{XX} (f_X) the XX (X) resonance frequency. Changing the cavity length δ_z allows τ_{XX}/τ_X to be tuned *in situ* over 2 orders of magnitude. The asymmetry in the $|XX\rangle$ lifetime in (a) comes from enhancement of the transition by the second cavity mode at higher positive detunings. (c) Schematic of the open microcavity enhancing $|XX\rangle \rightarrow |X\rangle$, and (d) enhancing $|X\rangle \rightarrow |0\rangle$.

dot-to-dot fluctuations in the lifetime ratio [23] or on tuning via an applied electric field [28]. In the first case, V was limited to a small range around 60%. In the second case, while V_{XX} could be increased to 76.9%, the electric field required to increase τ_X reduced V_X to 42%: the improvement in XX photon coherence thus came at the expense of X coherence, a clear departure from Eq. (1).

Here, we explore experimentally the dependence of V on lifetime ratio upon tuning τ_{XX}/τ_X over 2 orders of magnitude. In one extreme, $\tau_{XX}/\tau_X = 0.08$, we measure “raw” visibilities (not corrected for imperfect photon purity) $V_{\text{raw}}^{XX} = 90 \pm 2\%$ and $V_{\text{raw}}^X = 80 \pm 6\%$, improving the coherence for both XX and X photons. Conversely, in the other extreme, $\tau_{XX}/\tau_X = 6.2$, $V_{\text{raw}}^{XX} = 11 \pm 3\%$ ($V_{\text{raw}}^X = 15 \pm 1\%$ for $\tau_{XX}/\tau_X = 5.5$). As we show, these results closely match the theoretical prediction.

The experiment uses a self-assembled InGaAs QD in an open microcavity [11,29], Figs. 2(c) and 2(d). The bottom mirror is a high-reflectivity distributed Bragg reflector within the semiconductor heterostructure. The reflectivity of the top mirror, a curved micromirror fabricated in a silica substrate, is lower and is chosen to optimize the end-to-end efficiency [11]. The cavity has finesse 500 and Q factor 10,000. The QD charge is controlled by Coulomb blockade using an $n-i-p$ diode and the charge is set to zero in these experiments. The biexciton is excited by resonant two-photon excitation (TPE) using a 5-ps laser pulse tuned in

frequency to lie at the mean frequency of the X and XX transitions. Both XX and X photons are collected; their frequency separation (the biexciton binding energy) is 690 GHz. In the output, either XX or X photons are selected with a spectral filter, and the quantum optical properties [autocorrelation $g^{(2)}(0)$ and Hong-Ou-Mandel visibility V] are determined following standard protocols [30]. One potential complication is the exciton fine structure (2.6 GHz), likewise the mode splitting (50 GHz) of the fundamental cavity mode [11,37]. The mode splitting is sufficiently large that if one mode is close to resonance, then the other mode can be ignored. Moreover, we estimate that the exciton and cavity axes are misaligned by less than 5 deg. This is not coincidental: both are strongly influenced by the semiconductor crystal axes [37]. It is therefore safe to describe the experiments in terms of one cascade pathway and one cavity mode.

There are three key enabling features in these experiments. First, the cavity frequency can be tuned *in situ*, in practice by controlling the separation δz between the semiconductor heterostructure and the top mirror, Figs. 2(c) and 2(d). The cavity mode has a δz linewidth of 920 pm, and the root-mean-square noise in δz is much less, just 10 pm [38]. This allows us to tune precisely the cavity mode into resonance either with $|XX\rangle \rightarrow |X\rangle$ or $|X\rangle \rightarrow |0\rangle$ all on the same QD, hence to explore V over a large range of lifetime ratios. Second, the heterostructure operates with very little charge noise [7,11]. The transition linewidths are in all cases close to the transform limit and much smaller than the cavity linewidths. This means that the Purcell factors are not subject to noise. Also, the noise at frequencies above 100 kHz is almost nonexistent such that the HOM experiments, carried out here by interfering photons separated in time by 13.1 ns, are unaffected by the residual weak and slow noise processes. Third, cavity photons are coupled into a single-mode “collection” fiber with high efficiency, $\simeq 69\%$. This high cavity-to-fiber efficiency allows us to perform two-photon correlation measurements on X photons when the XX is coupled to the cavity (and vice versa) even though the out-of-resonance exciton couples very weakly to the cavity.

We first demonstrate control of the $|XX\rangle$ and $|X\rangle$ lifetimes via the Purcell effect. When $|XX\rangle \rightarrow |X\rangle$ ($|X\rangle \rightarrow |0\rangle$) is in resonance with the cavity mode we collect XX (X) photons efficiently and X (XX) photons very inefficiently. In the on-resonance case, the photons are emitted preferentially into the cavity mode from which they escape through the top mirror and propagate to the detector. In the out-of-resonance case, the photons are emitted preferentially into other modes. Nevertheless, a residual coupling to the cavity occurs via the frequency tails of the cavity mode despite the large difference between detuning (690 GHz) and cavity linewidth (25 GHz) [39]. This enables us to detect both XX and X photons for all cavity detunings [30]. Plotted in Figs. 2(a) and 2(b) are the

measured $|XX\rangle$ and $|X\rangle$ lifetimes as a function of cavity detuning [30]. There is a clear Purcell effect on both $|XX\rangle$ and $|X\rangle$. The Purcell factors are 6.9 and 11.2, respectively. The difference in Purcell factors originates from the two decay pathways for the $|XX\rangle$ compared to the single pathway for each of the $|X\rangle$ [30].

We proceed to measure $g^{(2)}(0)$ and HOM visibility for both XX and X as a function of cavity detuning. The experiments are straightforward on the transition resonant with the cavity but challenging on the transition out of resonance with the cavity. For instance, when X is resonant with the cavity, the β factor, which quantifies the fraction of photons emitted into the cavity mode, for X is 91% resulting in a large flux of X photons, yet XX is far detuned—the X-XX splitting is 24 times larger than the cavity linewidth—such that almost all XX photons are emitted into noncavity modes. (We estimate that the XX β factor is just 0.3% in this scenario.) The two-photon coincidence rates depend on β^2 : an out-of-resonance case requires a 10^5 -times longer integration time than an on-resonance case. The on-resonance $g^{(2)}(0)$ and HOM visibility are recorded in roughly a minute, such that out-of-resonance $g^{(2)}(0)$ and HOM measurements are feasible by integrating over about a day. Given this integration time, we sample the out-of-resonance cases only at the two extreme points (XX when X is resonant with the cavity, and vice versa). Both $g^{(2)}(0)$ and HOM visibility are determined by adding up all the counts in a time window covering > 16 lifetimes around delay zero [30]. In all cases, the HOM experiments measure V_{raw} , the “raw” visibility, Fig. 3. In general, two-photon emission events, i.e., a nonzero $g^{(2)}(0)$, contribute to V_{raw} . The two resonance cases behave differently. With X in resonance with the cavity, $g^{(2)}(0) < 0.1\%$, such that multiphoton emission events have a negligible impact on the HOM visibility V . With XX in resonance with the cavity, $g^{(2)}(0) = 2.3\%$ on account of cavity feeding through the $|X\rangle$, caused by phonon-mediated QD-cavity interactions. To estimate V in the limit $g^{(2)}(0) \rightarrow 0$, we use $V = [1 + 2g^{(2)}(0)]V_{\text{raw}}$ [40]. In the nonresonant cases, the coincidence rates are so low that a very weak background signal (arising from incoherent room light coupled into the fibers) contributes to the signal. Only in one case is the background relevant and it is subtracted to determine $g^{(2)}(0)$ and V_{raw} [30].

Figure 4 shows our central result, the dependence of V on lifetime ratio. The plot uses all the data, i.e., both XX and X for all cavity detunings. The results lie on a single curve, evidence for a general behavior, and follow Eq. (1). When the cavity is resonant with $|XX\rangle \rightarrow |X\rangle$, both XX and X photons have a high value of V , $94 \pm 2\%$ and $82 \pm 6\%$, respectively. Conversely, when the cavity is resonant with $|X\rangle \rightarrow |0\rangle$, both XX and X photons have a low value of V , $12 \pm 3\%$ and $15 \pm 1\%$, respectively. In the former case, $|XX\rangle$ decays much more rapidly than $|X\rangle$,

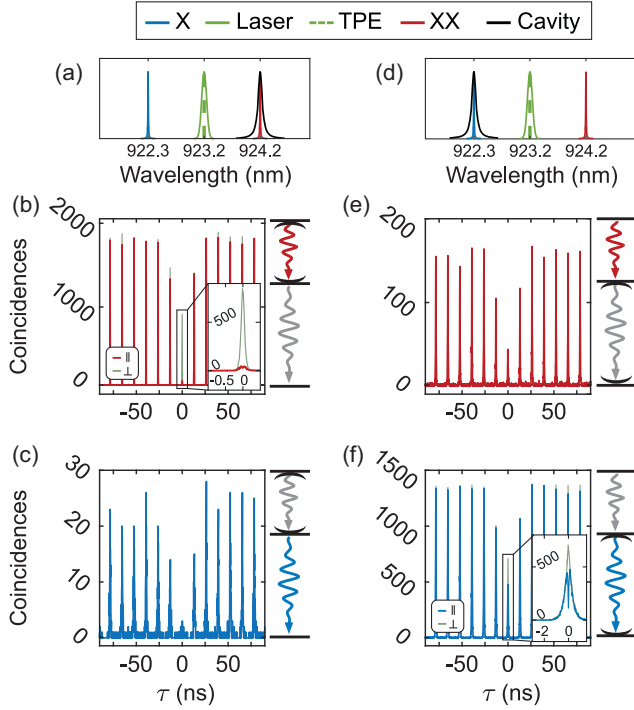


FIG. 3. Characterization of the photon coherence using two-photon interference. (a) Frequency alignment of the X, XX, TPE laser and cavity for Purcell enhancement of the XX transition to reduce τ_{XX} . This configuration improves the photon coherence for both XX and X photons. (b) Two-photon interference data for the XX photons, with $V_{\text{raw}}^{\text{XX}} = 90 \pm 2\%$. (c) Two-photon interference data for the X photons, with $V_{\text{raw}}^{\text{X}} = 80 \pm 6\%$. (d) Frequency alignment for Purcell enhancement of the X transition, which increases τ_{XX}/τ_X and thus reduces the photon coherence for both XX (e) and X (f) photons. In each case, only the photons in one of the cascade pathways are detected (via a linear polarizer with an axis matching the cavity axis). X (XX) photons are selected and XX (X) photons rejected by a spectral filter with bandwidth 30 GHz.

such that the timing jitter of X becomes less important and high values of V are achieved [41].

We turn now to $g^{(2)}(0)$. When $|XX\rangle \rightarrow |X\rangle$ ($|X\rangle \rightarrow |0\rangle$) is in resonance with the cavity, $g^{(2)}(0) = 2.3 \pm 0.2\%$ ($g^{(2)}(0) < 0.1\%$). This difference in $g^{(2)}(0)$ is strong evidence for cavity feeding. When $|XX\rangle \rightarrow |X\rangle$ is in resonance with the cavity, $|X\rangle \rightarrow |0\rangle$ is blue detuned by the biexciton binding energy (690 GHz) from the cavity. $|X\rangle$ decays with high probability by emitting a photon into a noncavity mode but with non-negligible probability by emitting a photon into the cavity and creating a phonon to conserve energy, i.e., by cavity feeding [41]. In the converse case when $|X\rangle \rightarrow |0\rangle$ is in resonance with the cavity, $|XX\rangle \rightarrow |X\rangle$ is far red detuned, such that cavity feeding via XX emission relies on the absorption of a phonon, a process which is suppressed via the temperature (thermal energy 90 GHz, X-XX splitting 690 GHz). For this reason, $g^{(2)}(0)$ is very small when X is resonant with the cavity.

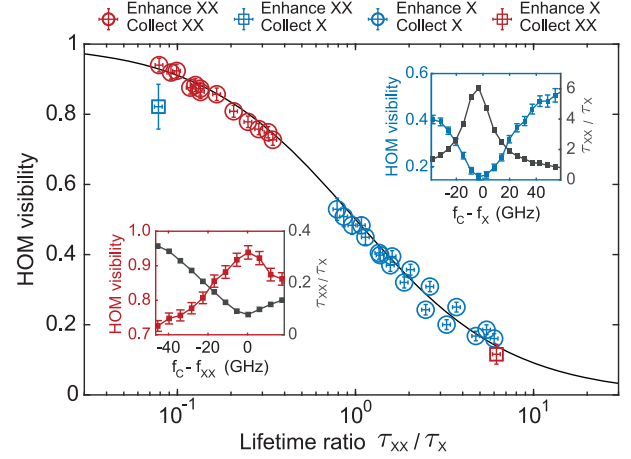


FIG. 4. Photon coherence V as a function of lifetime ratio τ_{XX}/τ_X . The points show $V = [1 + 2g^{(2)}(0)]V_{\text{raw}}$, where V_{raw} is the measured visibility after correcting for the imperfect beam splitter [30], and $1 - g^{(2)}(0)$ is the single photon purity. This correction removes the contribution of two-photon emission via cavity feeding from V and enables a fair comparison to the quantum optics theory, Eq. (1), solid line. Insets: HOM visibility and lifetime ratio as a function of cavity detuning from XX (red) and X (blue) transitions.

In quantum optics, Eq. (1) was derived using the Wigner-Weisskopf approach, i.e., emission into unstructured free space [24]. It is not *a priori* clear if the same result holds in our case with Purcell-enhanced emission into the cavity mode along with spontaneous emission into noncavity modes, here the one-dimensional continuum of surface modes at the GaAs-vacuum interface. To explore this, we apply cavity QED to the two-photon cascade [30]. In standard notation, κ is the photon decay rate out of the cavity, g the cavity field-QD coupling rate (assumed equal for XX and X), and $\gamma_{\text{bare}}^{\text{X}}$, $\gamma_{\text{bare}}^{\text{XX}}$ the decay rates into the one-dimensional continuum ($\gamma_{\text{bare}}^{\text{XX}} \simeq 2\gamma_{\text{bare}}^{\text{X}}$). We take the “fast-cavity” limit, $\kappa > g \gg \gamma_{\text{bare}}^{\text{X}}, \gamma_{\text{bare}}^{\text{XX}}$, appropriate to the experiments $[(\kappa, g, \gamma_{\text{bare}}^{\text{X}}) = (25, 4.4, 0.3) \times 2\pi \text{ GHz}]$. In this limit, we find that Eq. (1) holds even for a detuned cavity with $1/\tau_{\text{X,XX}}$ given by

$$1/\tau_{\text{X,XX}} = \gamma_{\text{Purcell}}^{\text{X,XX}}(\omega_{\text{cav}}) + \gamma_{\text{bare}}^{\text{X,XX}}. \quad (2)$$

The decay rates $\gamma_{\text{Purcell}}^{\text{X,XX}}$ describe the Purcell enhancement of X and XX (angular frequencies ω_X and ω_{XX} , respectively) via coupling to the same cavity mode (angular frequency ω_{cav}):

$$\gamma_{\text{Purcell}}^{\text{X,XX}}(\omega_{\text{cav}}) = \frac{g^2 \kappa}{(\omega_{\text{X,XX}} - \omega_{\text{cav}})^2 + (\frac{\kappa}{2})^2}. \quad (3)$$

In practice, the experiment measures τ_{XX} and τ_X . Hence, this analysis explains why V follows Eq. (1) as shown in Fig. 4.

As a final step, phonons can be included in the theory [41]. For cavity enhanced XX, V_X is predicted to be slightly lower than V_{XX} on account of phonon-induced dephasing of the relatively long-lived X. This is probably the explanation for the slightly reduced V_X at the smallest lifetime ratio in the experiment, Fig. 4.

To conclude, we have manipulated the coherence of the two photons in a two-photon cascade. When the upper transition (here the biexciton in a QD) is Purcell enhanced by a cavity, both photons become highly indistinguishable. Conversely, when the lower transition (here the exciton) is Purcell enhanced, both photons become distinguishable. The quantum optics result describes the experiments over two decades in lifetime ratio. Phonons in the semiconductor result in an effect beyond quantum optics: occasional emission of two cavity photons in one cascade via cavity feeding.

Our work demonstrates that the biexciton cascade is capable of producing photons with a high degree of indistinguishability. Cavity feeding will limit the indistinguishability (i.e., V_{raw}) in the limit of an ultrasmall XX:X lifetime ratio. It can be suppressed by using a QD with a very large XX binding energy [25], or by switching to larger QDs, for instance, GaAs QDs, for which the X-phonon coupling is smaller. To exploit these advances in a source of entangled photons in the polarization basis, it is necessary that precession caused by the fine structure splitting is much slower than X decay and there are established methods to do this [17]. In the time-bin basis, it is necessary to find a metastable state with an optical transition to the biexciton. In both schemes, it is also necessary to extract both XX and X photons efficiently. The open microcavity is a narrow band device and is not immediately suited to this task: if the XX photon enters the cavity mode, the X photon couples predominantly to the surface mode. Nevertheless, the X photons could be coupled out of the chip using an on-chip grating [42]. An alternative approach is to use a circular Bragg grating (bulls-eye) design for which the Purcell enhancement is wavelength selective yet the diffraction efficiency is high over a much higher spectral range [43,44]. The Purcell peak should be in resonance with the biexciton. Without Purcell enhancement, X decay will result in a weak phonon sideband. However, these red-shifted photons can be removed by a spectral filter and result mostly in a small reduction in efficiency. Our Letter shows that the biexciton decay can result in indistinguishability metrics above 90%.

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Data availability—The data presented in the main text are available via Zenodo at [45].

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