Abstract

We propose and theoretically analyze a new scheme for generating hyper-entangled photon pairs (EPPs) in a system of polaritons in coupled planar microcavities. Starting from a microscopic model, we evaluate the relevant parametric scattering processes and numerically simulate the phonon-induced noise background under continuous-wave excitation. Our results show that, compared to other polariton entanglement proposals, our scheme enables the generation of photon pairs that are entangled in both the path and polarization degrees of freedom, and simultaneously leads to a strong reduction in the photoluminescence noise background. This can significantly improve the fidelity of the EPPs under realistic experimental conditions.

Keywords: semiconductor microcavities, entanglement, polaritons, quantum photonics

1. Introduction

Entanglement is considered the primary resource for quantum information processing schemes and in the optical domain the practicability of implementing large-scale photonic quantum computers or long distance quantum communication protocols relies crucially on efficient...
sources for entangled photon pairs (EPPs). Conventionally, EPPs are produced by parametric down conversion in nonlinear crystals [1] or four-wave mixing in photonic crystal fibers [2]. Recently, the generation of EPPs using similar processes in microcavity polariton systems [3, 4] has been the subject of intensive research [5–10].

Being half exciton, half photon, polaritons benefit from strong Coulomb interactions, while they can easily be converted into propagating optical qubits for long-distance entanglement distribution. Potentially, this could be used to build sources with very high EPP generation rates (GRs). In addition the generation of entangled polaritons is an interesting goal in itself, especially in connection with ideas about polaritonic circuits [11]. However, the predicted quantum properties of generated polariton pairs [12–14] have remained quite elusive and no direct experimental evidence for entanglement detection has been demonstrated so far, mainly due to background noise caused by phonon-induced photoluminescence (PL) [15].

In this work we investigate the potential of coupled microcavity structures as bright sources of entangled and hyper-EPPs. In particular, we consider a system of three planar microcavities [7], which are coupled via two shared Bragg mirrors as illustrated in figure 1(a). In this setup the splitting of the upper and lower polariton dispersion curves into three well-separated sub-branches provides an additional flexibility for engineering parametric inter-branch scattering processes [13, 16], which leads to qualitative new features and an improved performance of the EPP creation. By pumping two polariton modes in the first and third branch, the phase matching condition for parametric scattering is fulfilled on an energetically degenerate circle of momentum states in the middle branch as shown in figure 1(c). By choosing different pumping configurations, photon pairs with polarization entanglement or pairs with entanglement in both the path and polarization degrees of freedom can be generated. Such hyper-entangled photons [17–19] provide a valuable resource for super-dense coding protocols [20, 21] or quantum key distribution [22, 23]. Moreover, the energy separation between the different branches creates a bottleneck for phonon-induced polariton scattering and under realistic conditions suppresses the background PL by several orders of magnitude. Our analysis of the resulting entanglement fidelity shows a significant improvement over previously analyzed single-cavity settings and that the proposed multi-cavity system offers a realistic approach for future solid state based EPP sources.

2. Model

In the setup shown in figure 1(a) the photons in each cavity are coupled to electronic interband excitations (excitons) of a semiconductor quantum well (QW) and form new quasi-particles—so-called polaritons—under strong coupling conditions. For the sake of simplicity in the notation, but without loss of generality, we are assuming one single QW inside each microcavity. When \( N \) identical QWs are used, only one ‘bright’ state will be coupled to the photon mode [24, 25] and the following Hamiltonian can still be used provided that the exciton modes in each cavity can be considered as these bright collective states. As a consequence the value of the Rabi light–matter interaction \( V_n \) can be tailored (to some extent) by design. We assume that both photons and excitons are restricted to a single mode function along the confined direction (\( z \)-axis) and we denote by \( E_{nk} \) (\( E_{nk}^* \)) the energy dispersion of photons (excitons) with transverse momentum \( k \) and cavity index \( n \). The Hamiltonian for the whole system is
\[ \hat{H} = \sum_{n,k} E_{nk} \hat{a}^\dagger_{nk} \hat{a}_{nk} + E_{nk} \hat{B}^\dagger_{nk} \hat{B}_{nk} + V_n \left( \hat{a}^\dagger_{nk} \hat{B}_{nk} + \text{h. c.} \right) \\
- \sum_{n,k} J_{n,n+1} \left( \hat{a}^\dagger_{nk} \hat{a}_{n+1k} + \text{h. c.} \right) + \hat{H}_C, \]

where \( \hat{a}_{nk} \) and \( \hat{a}_{nk}^\dagger \) are bosonic annihilation and creation operators for photons in cavity \( n \) and transverse momentum \( k \). \( \hat{B}_{nk} \) and \( \hat{B}_{nk}^\dagger \) are the corresponding operators for excitons. \( V_n \) is the dipole coupling strength between photons and excitons within cavity \( n \) and \( J_{n,n+1} \) is the optical mode coupling between neighboring cavities. Finally, \( \hat{H}_C \) accounts for the Coulomb interaction between excitons, which gives rise to the effective \( \chi^{(3)} \) polariton–polariton nonlinearity discussed below.

Many descriptions of polariton parametric processes make use of the picture of excitons as interacting bosons [8, 9, 15, 26, 27]. In these models bosonic commutation relations are assigned to the exciton operators \( \hat{B}_{nk} \) and \( \hat{B}_{nk}^\dagger \) and an effective boson–boson coupling is added to the Hamiltonian to account for the nonbosonic exciton character (‘phase-space filling’). Combined with the Coulomb interaction this coupling is then treated within the mean-field approximation.

**Figure 1.** (a) Setup. Three microcavities, each containing a quantum well (QW), are coupled via two distributed Bragg reflectors (DBR) [7]. Under cross-polarized laser excitation (right), the emitted signal and idler photons (left) are entangled in momentum and polarization degrees of freedom. (b) Energy dispersion curves of the six polariton branches (solid lines); the dashed lines show the energies of the bare exciton and the lowest photonic mode. (c) Actual lower polariton branches for the sample parameters discussed in the text. The red circles show the chosen pump wavevectors \( \left( k_p = -k_p', \, |k_p'| = 1.8 \mu m^{-1} \right) \) which, by mixed parametric scattering, generate a circle of energy-degenerate hyper-entangled signal-idler pairs (dashed blue line).
approximation. At low polariton densities this approach provides a good description of the resulting nonlinear third order response and is often used due to its simplicity and its direct analogy to nonlinear optics. However, in the resulting equations of motion the absolute strengths of the nonlinear terms differ from those obtained from more rigorous microscopic theories [28]. For completeness, these corrections are detailed in appendix A and all results presented here take them into account.

In the following we write \( \hat{H} = \hat{H}_0 + \hat{H}_{\text{int}} \), where apart from the Coulomb term, \( \hat{H}_{\text{int}} \) accounts for additional effective interactions due to the nonbosonic character of the excitons [8, 28]. The linear part, \( \hat{H}_0 \), can be diagonalized by a generalized Hopfield transformation \( \hat{P}_k = \sum_n X^n_k \hat{B}_{nk} + C^n_k \hat{a}_{nk} \), and expressed in terms of a set of polaritonic quasi-particle operators

\[
\hat{P}_k = \sum_n X^n_k \hat{B}_{nk} + C^n_k \hat{a}_{nk},
\]

where \( X^n_k \) and \( C^n_k \) are the exciton and photon components of the \( i \)th polariton branch. For a single cavity the index \( i \) denotes the familiar lower and upper polariton branches, which, for zero detuning, are split by \( \sim 2V \) around \( k = 0 \) [30, 31]. In coupled structures, the optical mode coupling induces a further splitting of \( \sim J \). Typical dispersion curves for three coupled cavities are shown in figure 1(b), where identical Rabi splittings \( 2V = 12 \text{ meV} \) [32, 33] and mirror couplings \( J = 4 \text{ meV} \) have been assumed.

3. Photon pair creation

The nonlinear optical response of our system is governed by the strong Coulomb interaction between excitons, \( \hat{H}_{\text{int}} \sim \hat{B} \hat{B} \hat{B} \hat{B} \), which results in an equivalent \( \chi^{(3)} \)-type nonlinearity between polaritons. In the following we are focusing on the lower set of branches \( i = 1, 2, 3 \), which have a smaller exciton–exciton dephasing [10, 14]. We assume that two polariton modes with wavevectors \( k_p \) and \( k'_p \) and energies \( E_{k_p} \) and \( E_{k'_p} \), respectively, are strongly pumped by external lasers and we linearize the interaction around the classical mean value of these two pumped modes. We obtain a parametric scattering process analogous to four-wave-mixing in nonlinear optical crystals,

\[
\hat{H}_{\chi^{(n)}} = \sum_{k, k'} \left( g P_{k_p}^* P_{k'_p}^* \hat{P}_{k_p} \hat{P}_{k'_p} + \text{h. c.} \right) \delta_{k, k', k + k'},
\]

where \( k = (i, \mathbf{k}) \) labels the polariton branch and the 2D wavevector. \( P_{k_p} \) and \( P_{k'_p} \) denote the classical amplitudes of the pumped modes and the interaction strength \( g \) depends on all the details of the four involved states\(^6\). Equation (3) describes the physical process of two coherent pump polaritons being scattered into a signal-idler polariton pair, which satisfies the phase matching conditions

\[^5\] This value can be achieved with some QWs in GaAs structures [32], or hybrid organic microcavities [33].

\[^6\] In particular, for signal-idler wavevector \( \mathbf{k}_p, \mathbf{k}_i \), the pump-mixed process in figure 1 has a nonlinear coefficient of the form \( g = \frac{1}{2} \sum_p \left( V/n_{\text{sat}} \right) X_{k_p k_i} X_{k_i k_p} X_{k_p k_i} X_{k_i k_p} \), with the saturation density \( n_{\text{sat}} \) and the Coulomb-induced contribution including its correction beyond mean-field \( V_{\text{c}} \).
\[
\begin{align*}
\begin{cases}
k_p + k' &= k_i + k_i, \\
E_{k_p} + E_{k'} &= E_{k_i} + E_{k_i}.
\end{cases}
\end{align*}
\]

The resulting shape of the available states depends on the energy dispersion curves and on the positions of the pump beams. In single planar cavity setups with one [15] or two pumps [14, 34] the available phase-space reduces to curves where at most two of the final states can have the same energy. Multi-mode settings provide a much larger flexibility [13]. For the present planar device and the pump configuration shown in figure 1(c), the phase-matching conditions (4) are fulfilled on a whole circle of energy-degenerate states in the middle polariton branch. This will allow simultaneously the generation of hyper-entangled states and the reduction of the detrimental phonon-induced noise background as shown below.

4. Polarization and path entanglement

For pump fields with a definite circular polarization ($\sigma = \pm 1$), this polarization will be inherited by the polariton modes and due to spin-preserving Coulomb interactions only one of the four polarization configurations $|+ + \rangle$, $|- - \rangle$, $|+ - \rangle$ or $|- + \rangle$ is created. For two linearly co-polarized pump beams all the four polarization states are activated and for the generation of polarization entanglement the only useful configuration occurs for linearly cross-polarized pump fields [14]. In this case the counter-circular channel (due to bound biexciton and two-exciton scattering states of opposite spin) is suppressed owing to destructive interference (see appendix B). As a result, the generated photon pairs are produced in an entangled state of the form $(|+ + \rangle + |- - \rangle)/\sqrt{2}$.

By generalizing equation (3) to account for polarization selection rules and restricted to cross-polarized driving, the photon pair creation process is described by an effective Hamiltonian

\[
\hat{H}_{\text{eff}} = \sum_{k,k'} G \left( \hat{P}^\dagger_{k_p} \hat{P}^\dagger_{k_p} + \hat{P}^\dagger_{k_p} \hat{P}^\dagger_{k_p} \right) + \text{h. c.},
\]

where $G = g \mathcal{P}_p \mathcal{P}_p$ and $k_i = k_p + k' - k_i$ is assumed. For the configuration shown in figure 1 c) $|k_i| = |k_p|$ and by selecting specific paths (say $k_1$ and $-k_1$ and $-k_2$) the outgoing photon pair is generated in a hyper-entangled state

\[
|\psi\rangle = \frac{1}{2} \left( |k_1, -k_1\rangle + |k_2, -k_2\rangle \right) \otimes \left( |+ + \rangle + |- - \rangle \right),
\]

which exhibits entanglement in both the momentum and the polarization degrees of freedom. Being degenerate in energy the photons in modes $k_1$ and $k_2$ can be interfered, and the momentum can be used as an independent degree of freedom in photonic entanglement experiments [18].

5. Photoluminescence

In polariton systems the fidelity of the EPPs is affected by phonon-induced scattering processes and Rayleigh scattering from the pump beams. The effect of Rayleigh scattering is strongly
suppressed as soon as the pump and the signal-idler photons are not degenerate in energy. In contrast, at temperatures of a few Kelvin, polaritons can scatter incoherently by emission or absorption of acoustic phonons, and redistribute along the dispersion curve. This pump-induced PL noise background competes with parametric coherent photoemission and lowers the degree of nonclassical correlations [34, 35]. The multi-cavity setting can reduce its impact by using the mode splitting $J$ to create a large energy separation between the pump and the signal-idler photons.

To quantify the amount of entanglement in the present coupled cavity setup, we study the competition between parametric coherent scattering and the incoherent PL background. Starting from equation (5), the dynamics of the signal and idler mode can be evaluated within a two-mode description [36]

$$\partial_t \hat{P}_{k_i} = -i \left( \omega_{k_i} - i \frac{\Gamma_{k_i}}{2} \right) \hat{P}_{k_i} - i G \hat{P}^\dagger_{k_i} + \hat{F}_{k_i}, \quad (7)$$

$$\partial_t \hat{P}^\dagger_{k_i} = i \left( \omega_{k_i} + i \frac{\Gamma_{k_i}}{2} \right) \hat{P}^\dagger_{k_i} + i G^* \hat{P}_{k_i} + \hat{F}^\dagger_{k_i}, \quad (8)$$

where the background PL is treated separately from the parametric interaction and enters through the time-dependent Langevin noise operators $\hat{F}_{k_i}$ and $\hat{F}^\dagger_{k_i}$. Under continuous driving conditions the effect of noise is fully determined by the stationary correlators $\Gamma_{F, F}^{\text{PL}}$ and $\Gamma_{F, F}^{\text{PL}}$ where the total polariton decay rates $\Gamma_{k_i}$ and the stationary occupancies $N_{k_i}^{\text{PL}}$ are evaluated in the following.

The dominant incoherent processes for optically generated polaritons in III-V systems are (acoustic) phonon-induced scattering and radiative losses. In strain-free heterostructures, the exciton–phonon interaction is well described by a 3D bulk-like model,

$$\hat{H}_{e-ph} = \sum_{k, q, n} \Xi(q, k) \hat{B}^\dagger_{n-k+q} \hat{B}_{n-k} (e^{i q \cdot \vec{r}} \hat{b}_{q} + h. c. ), \quad (9)$$

where $\hat{b}_{q}$ is the bosonic operator for phonons with transverse momentum $q$ and momentum $k$ along the confinement axis. The notation takes into account explicitly the separation between confined and free directions. Here a 'two-dimensional' exciton is written in terms of a three-dimensional envelope convolution (in direct or reciprocal space), $\vec{k} = (k, k_z)$, $\vec{r} = (x, z)$, $\rho = (x_e - x_h)$ for electron (e) and hole (h). The mass ratios read $\alpha_e = m_e / M$, $\alpha_h = m_h / M$, with $M = m_e + m_h$. In equation (9) $\lambda_n = n z_{qw}$ accounts for the position of the wells numbered as $n = -1, 0, 1$ and the coefficient $\Xi(q, q_e)$ contains the exciton overlap integrals in terms of the Fourier transform over the phonon wave vectors [36, 38]. It reads

$$\Xi(q, q_e) = \left( \frac{\hbar q}{2 du V} \right)^{1/2} \left( D_{I_e} \left( q_e \right) I_{e} \left( q \right) + D_{I_h} \left( q_e \right) I_{h} \left( q \right) \right). \quad (10)$$

where $V = SL$ is the quantization volume, $L$ is the confined direction quantization length and $S$ is the quantization surface in the free directions; $u$ is the sound velocity, $d$ is the material density, $q = |\vec{q}|$ is the modulus of the 3D wave vector $\vec{q}$, while $D_e$ and $D_h$ are deformation
potential constants for the conduction and valence band, respectively. The two overlap integrals are given by

$$\int \chi_e(z) \chi_h(z) \, dz = \int d^2 \rho \, W(\rho) \int d^2 q \, e^{i \mathbf{q} \cdot \mathbf{e}} \rho_\parallel \cdot \mathbf{e} \left( 1 + \left( \frac{m_\parallel(c)}{2M} q a_s \right)^2 \right)^{-3/2}.$$  

Equation (11)

The second integral in the plane can be explicitly calculated owing to the form of the 1S exciton wavefunction, $\rho = \pi^2 - \rho_\parallel$, while $\chi_e(z)$ are the electron (hole) QW confined wave functions along the growth direction $z$.

Using Fermi’s golden rule, the linear PL dynamics for the populations $N_{kPL}^{\sigma}$, for each $\sigma$, can be described microscopically by a Boltzmann equation

$$\frac{\partial N_{k\sigma}^{PL}}{\partial t} = I_k - I_k^{\sigma} N_{k\sigma}^{PL} + \sum_k W_{k,k'}^{\sigma} N_{k'}^{\sigma},$$  

Equation (13)

where the total linewidth $I_k = I_k^{\sigma} + \gamma_k^{\text{rad}}$ includes phonon-induced and radiative losses. Under continuous wave (CW) excitation with two lasers resonant with $E_{k_p}$ and $E_{k_p'}$, we model the pump term $I_k$ as two Gaussian profiles centered around $k_p$ and $k_p'$ (see appendix C). The total phonon-induced polariton scattering rate from state $k$ to state $k'$ is $W_{k,k'}^{\sigma} = W_{k,k'}^{\sigma} + W_{k,k'}^{\sigma}$, where the rates from phonon emission (+) and absorption (−) are given by

$$W_{k,k'}^{\sigma} = \frac{1}{\rho u s} \frac{|\mathbf{k}' - \mathbf{k}|^2 + (q_z^0)^2}{\hbar u q_z^0} \left| \Xi (\mathbf{k} - \mathbf{k}', q_z) \right|^2 \times \left( n_B (E_{\bar{q}}^{\text{ph}}) + \frac{1}{2} \pm \frac{1}{2} \right).$$  

Equation (14)

Here $n_B (E)$ is the Bose distribution. In equation (14) $E_{\bar{q}}^{\text{ph}}$ is the phonon energy and the 3D phonon wave vector is $\bar{q} = (q = k - k', q_z^0)$, where $q_z^0$ is calculated from the condition of energy conservation, $\hbar \omega_k - \hbar \omega_{k'} \pm E_{\bar{q}}^{\text{ph}} = 0$. The sum over all final scattering states gives the total phonon-induced loss rate $I_k^{\text{ph}} = \sum_{k'} W_{k,k'}^{\text{ph}}$. The radiative linewidth is $\gamma_k^{\text{rad}} = \sum_n |X^n_k|^2 \gamma^n_c$, and in the following we take a typical cavity loss rate of $\gamma_c = 0.35$ meV.

Figure 2 shows the stationary polariton occupations $N_{k\sigma}^{PL}$ derived from equation (13) and for other parameters specified above and in [15]. For simplicity, but without loss of generality, radial symmetry is assumed (see appendix C) and the population distribution is plotted as a function of $|k|$. This is consistent with radial-symmetric steady-state populations observed in experiments [15, 37]. Phonon-induced polariton scattering favors quasi-elastic events exchanging small energies [38]. If more branches are present the intra- and inter-branch scattering rates will be of the same order of magnitude and the polariton population will spread across the branches as well. The pumped polariton modes in the first and third branch scatter...
dominantly with low energy exchanges into the second branch, which is separated from the other branches by more than 2 meV. Since under the relevant conditions $\gamma_k^{(\text{rad})} \gg \Gamma_k^{(\text{ph})}$ multi-phonon processes are highly suppressed, this creates a PL window for the signal and idler wave vectors around $k = -1.36 \, \mu m^{-1}$, where the PL population is reduced by more than three orders of magnitude with respect to the intra-branch PL on the third branch.

6. Entanglement quantification

To provide a direct comparison between the present multi-mode setup and entanglement generation schemes using only a single microcavity, we evaluate in figure 3 three different quantities that are of importance for the experimental verification of entanglement in these systems. The previously considered standard configuration for the single-cavity case is that of a single pump near the inflection point of the lower polariton branch (the so-called ‘magic angle’ $k_m$) giving rise to a signal/idler pair at $k_s = 0$ and $k_i = 2k_m$ respectively. This configuration suffers from the fact that the two generated polaritons will have very different energies and photonic components. This imbalance spoils quantum correlations and the detection and manipulation of entangled photons at very different frequencies is challenging. In order to provide a fair comparison, we shall consider the balanced situation of [14] where a two-pump scheme below the magic angle creates balanced signal and idler pairs at a finite wavevector (see [14] for details).

In figure 3(a) we first plot the signal to noise ratio (SN) defined as the total generated signal $N_s$ over the background PL $N_{PL}$ at the detected signal and idler wavevectors. The red line shows the results obtained from the steady state solution of equations (7) and (8) and the PL noise level evaluated with equation (13). The black line indicates the corresponding results for a single-cavity setup [14], but with otherwise identical parameters. We see that for a single cavity, the emitted photons are dominated by the PL background, whereas in the three-cavity setting the
coherent signal is clearly above the PL noise level and dominated by the parametric (pair) emission.

Under the same conditions we evaluate in figures 3(b) and (c) the equal time two-photon correlation function $g^{(2)}(0) = \left\langle \hat{P}_a^\dagger \hat{P}_a^\dagger \hat{P}_b \hat{P}_b \right\rangle / \left\langle \hat{P}_a^\dagger \hat{P}_a \right\rangle \left\langle \hat{P}_b \hat{P}_b \right\rangle$ and the entanglement of formation (EOF) [39] as two measures of signal-idler correlations and of the degree of polarization entanglement [40, 41] between two simultaneously emitted photons, respectively. The EOF is evaluated from the reconstructed two-polariton subsector of the full density operator [42, 43], which can be obtained experimentally from measurements of all the four-operator expectation values $\left\langle \hat{P}_a(t_1) \hat{P}_a^\dagger(t_2) \hat{P}_b(t_3) \hat{P}_b^\dagger(t_4) \right\rangle$. In contrast to the total signal, the quantities plotted in figures 3(b) and (c) are sensitive to detected photon pairs only and consistent with previous findings [14] we observe bunching, $g^{(2)}(0) > 1$, and polarization entanglement EOF > 0 for both configurations. However, the bunching of $g^{(2)}(0) \sim 4$ achievable with the single-cavity set-up is at the limit of the acquisition capabilities of current experiments [40], while the triple
cavity structure shows a ten-fold enhanced effect, $g^{(2)}(0) > 30$, in relation with a very high SN ratio. Moreover, there is a pronounced difference in the fidelity of the EPP, and the EOF in the three-cavity setting can be close to the value of $\text{EOF} = 1$ expected for a pure polarization entangled Bell state.

7. Estimation of the brightness of our source

For a standard detection scheme, the far-field real space image obtained through a typical system of lenses has a total area of about $5 \text{ mm} \times 5 \text{ mm}$ corresponding to a grid spacing in reciprocal space of about $5 \mu\text{m}^{-1}$. By selecting photons of specific $k$-vector through a pinhole or optical fibers we can obtain a resolution of about $50 \mu\text{m}$, which equals an area of $A_{\Delta k} = (\Delta k)^2 = (0.05)^2 \mu\text{m}^{-2}$ in momentum space [15]. This area is much smaller than the momentum uncertainty from the polariton loss rate, such that we can approximate the arc of the circle with its tangent straight line. Moreover, $\Delta k$ is so small with respect to the thickness of the generated circle (proportional to the polariton loss rate $\Gamma$) that we can actually consider the momentum spread of the signal population in the perpendicular direction as constant. The actual sample size of the microcavity can be tens of mm, while, in contrast, the spot size of our Gaussian beam in real space is about $\sigma \approx 10–20 \mu\text{m}$, which largely covers the region where polariton and parametric processes are produced, giving a fairly homogeneous excitation. As a consequence, in our estimation we can assume that everything happens, as if the sample size was determined by the area of the laser spot $A_L = \pi\sigma^2$. Over the resolution spot $A_{\Delta k}$ the generated signal population is mostly constant, and then the number of detected photons will be proportional to the number of $k$ modes inside the detection area, scaling as $A_{\Delta k} A_L$.

The GR of correlated photon pairs can then be estimated as

$$\text{GR} = \frac{1}{\hbar} \left( A_{\Delta k} A_L \right) \frac{\Gamma}{h} \frac{N_k}{N_s} \simeq \frac{\Gamma}{h} \frac{N_k}{N_s} 1.76 . \quad (15)$$

Plugging in the values we obtain from the simulation, we estimate the brightness of our source in a range of sensible pump powers as $\text{GR} (I = 10 I_0) \approx 5 \times 10^8 \text{ s}^{-1}$, $\text{GR} (I = 20 I_0) \approx 1.5 \times 10^9 \text{ s}^{-1}$, with the reference pump intensity $I_0$ corresponding to an injected polariton density of about $0.5 \mu\text{m}^{-2}$ [15]. Under the same conditions ($I = 20 I_0$) the total EPP production rate GR can be significantly higher than what is currently achieved with conventional EPP sources [44].

8. Conclusions

In this work we showed that coupled microcavity structures can become an innovative design for protecting quantum coherences from solid-state background noise, and we applied this concept to devise novel bright sources of entangled and hyper-EPPs. Our analysis predicts a suppression of the phonon-induced noise of more than three orders of magnitude, which thereby eliminates one of the main noise sources in current experiments.
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Appendix A. Polariton $\chi^{(3)}$-nonlinearity

The most popular description of polariton–polariton interaction is that of interacting bosons. In these theories, bosonic commutation rules are applied to excitons and a phenomenological interaction model Hamiltonian is postulated such as to reproduce the form of the nonlinear equation of motion [8, 9, 15, 26, 27]. At low polariton densities, this approach provides a good description of the linear dynamics and it is often applied to nonlinear third-order interaction within a mean-field approximation thanks to its simplicity and direct analogy to nonlinear optics. However, even if the resulting equations of motion shares the same form of those derived from microscopic descriptions, the absolute strengths of the nonlinear terms differ and one is lead to question their validity when competition among different scattering mechanisms (like polariton–polariton scattering versus phonon-mediated decoherence) becomes important. In this appendix we will generalize the arguments of [28] to the case of coupled cavities and give an intuitive, yet rigorous, account of the microscopic corrections.

A.1. Heisenberg equations of motion

In the case of a single cavity and a single pump field, the Heisenberg equations of motion for a polariton system up to the third-order nonlinearity have been fully worked out in [28, 36]. Taking these references as our starting point, we adapt here this analysis for the case of multiple coupled cavities. For concreteness we address directly the triple cavity case ($n_{\text{cav}} = 3$), but the following line of argument is completely general. We assume that the first cavity is driven by two coherent pump fields, which resonantly excite the polariton modes with wave vectors $k_p$ and $k'_p$, respectively. Following the same notation as in the paper, we introduce a vector $\mathbf{B}_k$ with components

$$\begin{pmatrix} \hat{B}_{1k} & \hat{a}_{1k} & \hat{B}_{2k} & \hat{a}_{2k} & \hat{B}_{3k} & \hat{a}_{3k} \end{pmatrix}^T$$

and write the resulting equations of motion in a compact form as

$$\dot{\mathbf{B}}_k = -i\Omega_{k}^{\text{sc}} \mathbf{B}_k + \mathbf{E}_k^{\text{in}} - i\mathbf{R}_k^{NL}.$$  \hfill (A.1)

Here the matrix

$$\Omega_{k}^{\text{sc}} \equiv \begin{pmatrix} \omega_{1k} & V_1 & -J_{1,2} & 0 & 0 & 0 \\ V_1 & \omega_{1k} & -J_{1,2} & V_2 & 0 & 0 \\ -J_{1,2} & V_2 & \omega_{2k} & 0 & -J_{2,3} & 0 \\ 0 & 0 & -J_{2,3} & V_3 & 0 & 0 \\ 0 & 0 & 0 & 0 & \omega_{3k} & -J_{1,2} \\ 0 & 0 & 0 & 0 & -J_{1,2} & \omega_{3k} \end{pmatrix},$$ \hfill (A.2)
describes the linear dynamics of the coupled modes and the vector $E_k^{in}$ accounts for the external driving field. For the pump configuration specified above

$$\begin{align*}
    E_k^{in} &= t_1 \left( E_{p} e^{-i\omega t} \delta_{k,k'} + E_{p} e^{-i\omega t} \delta_{k,k'} \right) \\
    E_k^{in} &= 0 \quad \text{otherwise.}
\end{align*}$$

(A.3)

The last term in equation (A.1), $R_k^{NL} = \left( \hat{R}_k^{NL}, 0, \hat{R}_k^{NL}, 0, \hat{R}_k^{NL}, 0 \right)^T$, describes additional nonlinear contributions, which we address in the following. Note that in equation (A.1) we have for simplicity omitted the photon loss terms, which can be included by adding photon loss rates for the cavity operators $\hat{a}_{nk}$ and the corresponding Langevin noise operators.

Coulomb and photon–exciton interactions will be effective only between exciton and photon mode belonging to the same cavity. As a consequence the relevant nonlinear source terms, which couple waves with different in-plane wave vector $k$, can be treated very similarly to the single cavity case. We write the nonlinear terms as

$$\begin{align*}
    \hat{R}_{nork}^{sat} &= \sum_{\sigma_{nk}'} \hat{B}_{nork} \hat{a}_{nork} \hat{B}_{nork}^\dagger, \\
    \hat{R}_{nork}^{xx} &= \int_{-\infty}^{t} \int_{-\infty}^{t'} \hat{T}^{++}(t-t') \hat{B}_{nork}(t') \hat{B}_{nork'}(t') \hat{B}_{nork}(t') \hat{B}_{nork'}(t').
\end{align*}$$

(A.4)

where $n_{sat} = 7/(16\pi a_k^2)$ is the exciton saturation density [28, 45] and $\hat{k} = k' + k'' - k$. The second term comes from the Coulomb interaction among electrons and holes and contains two contributions. In the co-circular channel, only particles with the same spin are involved and the resulting interaction is always repulsive. In contrast, the counter-circular term describes the scattering of excitons of opposite spin and it can include a bound biexciton intermediate state. As a consequence its strength and its sign can vary considerably around the biexciton binding energy [46]. Altogether, and using the same notation as in [46], we obtain

$$\begin{align*}
    \hat{R}_{nork}^{xx}(t) &= \int_{-\infty}^{t} \int_{-\infty}^{t'} \hat{T}^{++}(t-t') \hat{B}_{nork}(t') \hat{B}_{nork'}(t') \hat{B}_{nork}(t') \hat{B}_{nork'}(t').
\end{align*}$$

The transition T-matrix includes the instantaneous mean-field exciton–exciton interaction contribution and a noninstantaneous term originating from four-particle correlations [28, 45, 46]. The memory-less equal-time limit of equation (A.5) corresponds to an energy independent $\chi^{(3)}$ nonlinearity for excitons, which is usually assumed in effective models, where excitons are treated as interacting bosons. As we show below, within our microscopic theory and in a quasi stationary regime the resulting $\chi^{(3)}$ interaction between polaritons can be derived more rigorously by using a Wigner–Weisskopf approximation for the integral kernel.
A.2. Strong coupling and polariton–polariton interactions

When the coupling rate $V$ exceeds the decay rate of the exciton coherence and of the cavity field, the system enters the strong coupling regime. In this regime, cavity-polaritons arise as the two-dimensional eigenstates of $\Omega^\infty_{k}$. 

In order to obtain the dynamics for the polariton system we diagonalize the linear subproblem of equation (A.1) by a (unitary) Hopfield transformation [29] $\mathbf{P}_k = \mathbf{U}_k \mathbf{B}_k$, where $(\mathbf{P}_k)_i = \hat{P}_{ik}$, and express excitons and photons in terms of a set of polaritonic quasi-particle operators

$$\hat{P}_i = \sum_n X_{ik}^n \hat{B}_{nk} + C_{ik}^n \hat{a}_{nk},$$

where $X_{ik}^n$ and $C_{ik}^n$ can be seen as the exciton and photon components of the $n$th cavity on the $i$th polariton branch. By applying this transformation to equation (A.1) we obtain

$$\omega \partial_{\tau} \hat{P}_i = - i \omega_{ik} \mathbf{P}_i + \hat{E}_{ik}^{\text{in}} - i \hat{\mathbf{R}}^\text{NL}_{ik},$$

where the $\omega_{ik}$ are the polariton eigenfrequencies, $\hat{\mathbf{R}}^\text{NL}_{ik} = \mathbf{U}_k \mathbf{R}^\text{NL}_{ik}$ and $\hat{E}_{ik}^{\text{in}} = \mathbf{U}_k \mathbf{E}_k^{\text{in}}$.

Let us consider a situation where the energies of the exciting pulses are all close to the corresponding polariton resonance values $\omega_{ik}$ and the broadenings are small compared to the splitting between the polariton branches. By adopting a Wigner–Weisskopf approximation [45] we can simplify the memory integral in equation (A.5) in the case of CW excitation and express the result in terms of polariton operators $\hat{P}_{ik}$. The nonlinear interaction terms we obtain are given by

$$\left( \mathbf{R}^\text{NL}_{ik} \right)_i = \sum_{n=1}^{3} \left( \left( \mathbf{R}^\text{sat}_{ink} \right)_i + \left( \mathbf{R}^{xx}_{ink} \right)_i \right),$$

with

$$\left( \mathbf{R}^\text{sat}_{ink} \right)_i = \sum_{k,k',l,m,r} \hat{P}^{\uparrow}_{mok} (t) \left[ X_{ik}^n X_{mk}^n \frac{V}{\hbar} X_{nk}^n C_{nk}^n \right] \hat{P}_{lok} (t) \hat{P}_{nk} (t)$$

and

$$\left( \mathbf{R}^{xx}_{mk} \right)_i = \sum_{k,k',l,m,r} \hat{P}^{\uparrow}_{mok} (t) \left[ X_{ik}^n X_{mk}^n T^{++} (\omega_{ik} + \omega_{k}) X_{rk}^n X_{rk}^n \right] \hat{P}_{lok} (t) \hat{P}_{nk} (t) +$$

$$+ \sum_{k,k',l,m,r} \hat{P}^{\downarrow}_{mok} (t) \left[ X_{ik}^n X_{mk}^n T^{+-} (\omega_{ik} + \omega_{k}) X_{rk}^n X_{rk}^n \right] \hat{P}_{lok} (t) \hat{P}_{nk} (t),$$

where $l, m, r$ refer to polariton branches and $T^s (\Omega) = \int_{-\infty}^{\infty} \tilde{T}^s (\tau) e^{-i\Omega\tau} d\tau$ is the Fourier transform of the time-dependent kernel in equation (A.5) for $s = ++, +-, --$. This approach, which is valid under quasi-stationary conditions, fully accounts for the energy dependence of the exciton–exciton scattering by including in the above equations the frequency dependence of the $T$-matrix. It can be shown [45] that the co-circular channel can be written as the sum of a constant mean-field term $V_{xx}$ plus a genuine four-particle contribution, $T^{++} (\omega) = V_{xx} + F (\omega)$. In the present work we are interested in CW driving and in the scattering of polaritons with a pre-specified energy. In this case the frequency dependence of $T^{++} (\omega)$ simply leads to a renormalization of the coherent scattering amplitude. In order to maintain a connection with the
literature, in the following we will use loosely $V_{xx}$ as the Coulomb-induced co-circular interaction including its correction beyond mean-field.

In the last step we now assume that the two pumped polariton modes are strongly driven and we replace the corresponding operators by their classical expectations values, $\hat{P}_{k_p} \to \mathcal{P}_{k_p} = \langle \hat{P}_{k_p} \rangle$ and $\hat{P}_{-k_p} \to \mathcal{P}_{-k_p} = \langle \hat{P}_{-k_p} \rangle$, where again the convention $k_p = (i_p, k_p, \sigma_p)$ is assumed. As discussed below, we are mainly interested in cross-polarized pump beams to get rid of the spin nonconserving scattering channel by destructive interference. However, for the moment we will leave the polarization of the pump fields unspecified. Then, by retaining only the most relevant contribution $\sim \mathcal{P}_{k_p} \mathcal{P}_{k_p}$ in the expressions given in equations (A.9), the equation of motion for a generic polariton operator reads

$$\frac{d}{dt} \hat{P}_{ik} = -i \omega_k \hat{P}_{ik} - i \sum_{\sigma_p, \sigma_{-p}, \theta} \hat{P}_{\sigma_p \sigma_{-p} \theta} \left( g_{ik} \mathcal{P}_{i_p \sigma_p \theta} \mathcal{P}_{-i_p \sigma_{-p} \theta} \right), \tag{A.10}$$

where $g_{ik}$ is a short notation for the coupling strength, which depends on all the $\sigma$’s and $k$’s in equation (A.10) and is detailed below. This equation of motion can also be obtained in the Heisenberg picture by the effective Hamiltonian $H_{eff}$ (equation (3) in the paper). The branch index $m$ and wave vector $\vec{k}$ are defined by the wave vector and energy conservation rules,

$$\begin{cases}
k_p + k_{-p} = \vec{k} + \vec{k} \\
E_{i_p, k_p} + E_{-i_p, -k_p} = E_{i, \vec{k}} + E_{m, \vec{k}}.
\end{cases} \tag{A.11}$$

The nonlinear coupling strength can be written as the sum of the co-circular and counter-circular terms, $g_{ik} = g_{ik}^{++, \sigma} + g_{ik}^{++, \sigma}$, and it includes all the spin selection rules. They read

$$g_{ik}^{++, \sigma} = \sum_{n=1}^{n_{sat}} \left( \delta_{\sigma, \sigma_{-p}} \delta_{\sigma_{-p}, \sigma} X_{ik \sigma} X_{\sigma \sigma_{-p}} \frac{V}{n_{sat}} \left( X_{i_p \sigma_p \sigma_{-p}} C_{i_p \sigma_{-p}} + X_{i_p \sigma_{-p} \sigma} C_{i_p \sigma} \right) + 2 T^{++} X_{ik \sigma} X_{\sigma \sigma_{-p}} \right),$$

$$g_{ik}^{++, \sigma} = \sum_{n=1}^{n_{sat}} \left( \delta_{\sigma, \sigma_{-p}} \delta_{\sigma_{-p}, \sigma} X_{ik \sigma} X_{\sigma \sigma_{-p}} 2 T^{++} X_{ik \sigma} X_{\sigma \sigma_{-p}} \right). \tag{A.12}$$

Appendix B. Perpendicular polarization: spin-conserving channels

As the calculation of [46] shows, the $++$ kernel in the four-particle nonlinear response does not become negligible even in a very negatively detuned region, but it retains a value of approximately one third of the $++$ channel. However, with two linearly cross-polarized pumps, we can show that the counter-circular channel (due to bound biexciton and two-exciton scattering states of opposite spin) is suppressed owing to destructive interference.

We choose two pump fields with polarization vectors $\hat{e}_p = \theta$ and $\hat{e}_{-p} = \theta + \pi/2$ (e.g. $\theta = 0$ means the first pump polarized along $\hat{x}$, while the second one along $\hat{y}$). From the equation for the pumped polariton mode reading
\[
\frac{d}{dt} \mathcal{P}_{\sigma \upsilon \mathbf{k}_p} = -i \left( \omega_{\mathbf{k}_p} - \frac{i \Gamma_p}{2} \right) \mathcal{P}_{\sigma \upsilon \mathbf{k}_p} + t C_{\mathbf{k}_p}^\mathbf{n} E_{\sigma \upsilon \mathbf{k}_p}^{(+)} (\hat{e}_p \cdot \hat{\sigma}),
\]

where \( \hat{e}_p = \cos \theta |x\rangle + \sin \theta |y\rangle = 2^{-1/2} (e^{i\theta} |+\rangle + e^{-i\theta} |-\rangle) \), we can see that any linearly polarized pump excites two circularly polarized polariton modes,

\[
\begin{align*}
\mathcal{P}^{(+)}_1(t) &= \mathcal{P}_{\mathbf{k}_p}(t) \frac{e^{-i\theta}}{\sqrt{2}}, \\
\mathcal{P}^{(+)}_2(t) &= \mathcal{P}_{\mathbf{k}_p}(t) \frac{e^{-i\theta}}{\sqrt{2}} e^{i\pi/2}, \\
\mathcal{P}^{(-)}_1(t) &= \mathcal{P}_{\mathbf{k}_p}(t) \frac{e^{i\theta}}{\sqrt{2}}, \\
\mathcal{P}^{(-)}_2(t) &= \mathcal{P}_{\mathbf{k}_p}(t) \frac{e^{i\theta}}{\sqrt{2}} e^{i\pi/2}.
\end{align*}
\]

By working out explicitly the spin sum in equation (A.10) we obtain

\[
\begin{align*}
\frac{d}{dt} \hat{\rho}_{\text{ork}} &= -i \omega_{\mathbf{k}_p} \hat{\rho}_{\text{ork}} - i g_{\text{ork}} \hat{\rho}_{\text{ork}}^\dagger \left( \mathcal{P}^{(+)}_1 + \mathcal{P}^{(-)}_1 \right) \left( \mathcal{P}^{(+)}_2 + \mathcal{P}^{(-)}_2 \right) = \\
&= -i \omega_{\mathbf{k}_p} \hat{\rho}_{\text{ork}} - i \hat{\rho}_{\text{ork}}^\dagger \left[ g_{\text{ork}}^{++} \left( \mathcal{P}^{(+)}_1(t) \mathcal{P}^{(+)}_2(t) \delta_{\sigma,+} + \mathcal{P}^{(-)}_1(t) \mathcal{P}^{(-)}_2(t) \delta_{\sigma,-} \right) \right. \\
&\left. + g_{\text{ork}}^{--} \left( \mathcal{P}^{(-)}_1(t) \mathcal{P}^{(+)}_2(t) + \mathcal{P}^{(-)}_1(t) \mathcal{P}^{(-)}_2(t) \right) \delta_{\sigma,-} \right].
\end{align*}
\]

The counter-circular term cancels out by destructive interference,

\[
\begin{align*}
\left( \mathcal{P}^{(-)}_1(t) \mathcal{P}^{(+)}_2(t) + \mathcal{P}^{(+)}_1(t) \mathcal{P}^{(-)}_2(t) \right) \delta_{\sigma,-} &= \\
&= \left( -i \mathcal{P}_{\mathbf{k}_p}(t) \frac{e^{i\theta}}{\sqrt{2}} \mathcal{P}_{\mathbf{k}_p}(t) \frac{e^{-i\theta}}{\sqrt{2}} + i \mathcal{P}_{\mathbf{k}_p}(t) \frac{e^{-i\theta}}{\sqrt{2}} \mathcal{P}_{\mathbf{k}_p}(t) \frac{e^{i\theta}}{\sqrt{2}} \right) \delta_{\sigma,-} = 0,
\end{align*}
\]

while the co-circular term reads

\[
\begin{align*}
&= \mathcal{P}^{(+)}_1(t) \mathcal{P}^{(+)}_2(t) \delta_{\sigma,+} + \mathcal{P}^{(-)}_1(t) \mathcal{P}^{(-)}_2(t) \delta_{\sigma,-} = \\
&= -i \frac{e^{-2i\theta}}{2} \left[ \mathcal{P}_{\mathbf{k}_p}(t) \mathcal{P}_{\mathbf{k}_p}(t) \delta_{\sigma,+} - \mathcal{P}_{\mathbf{k}_p}(t) \mathcal{P}_{\mathbf{k}_p}(t) e^{i4\theta} \delta_{\sigma,-} \right].
\end{align*}
\]

Choosing the first pump such that \( \theta = \pi/4 \) and neglecting an overall phase, equation (B.2) becomes

\[
\frac{d}{dt} \hat{\rho}_{\text{ork}} = -i \omega_{\mathbf{k}_p} \hat{\rho}_{\text{ork}} - i G \hat{\rho}_{\text{ork}}^\dagger \left( \delta_{\sigma,+} + \delta_{\sigma,-} \right)
\]

with \( G = g \mathcal{P}_{\mathbf{k}_p}(t) \mathcal{P}_{\mathbf{k}_p}(t) \), and \( g = g_{\text{ork}}^{++} / 2 \). This equation of motion can be obtained by the effective Hamiltonian equation (5) and ideally generates a pure entangled triplet state of the form

\[
|\Psi\rangle = \frac{1}{\sqrt{2}} (|+,-,-\rangle + |-,+,+\rangle).
\]
Appendix C. Numerical simulation of the Boltzmann equation

Using Fermi’s golden rule, the linear PL dynamics for the populations $N_{PL}$ can be described microscopically by a Boltzmann equation

$$\partial_t N_{ik}^{PL} = -\Gamma_{ik} N_{ik}^{PL} + I_{ik} + \sum_{lk} W_{iklk}^{(ph)} N_{lk}^{PL}, \quad (C.1)$$

where the total linewidth $\Gamma_{ik} = \Gamma_{ik}^{(ph)} + \gamma_{ik}^{(rad)}$ includes phonon-induced and radiative losses, which are independent of spin, and the scattering rates $W_{iklk}^{(ph)}$ are defined in the main text. For simulating the external driving field at a specific $k$ we take a Gaussian pulse centered around a wave vector $k_p$,

$$I_{ik} = I_0 e^{-\frac{(k-k_p)^2}{2\sigma^2}}. \quad (C.2)$$

The resulting coherent part of the population is then given by

$$\partial_t N_{ik}^{PL} \bigg|_{coh} = -\Gamma_{ik} N_{ik}^{PL} \bigg|_{coh} + I_{ik}. \quad (C.3)$$

In the paper we are mainly interested in the (incoherent) background PL defined as $\tilde{N}_{ik}^{PL} = N_{ik}^{PL} - N_{ik}^{PL} \bigg|_{coh}$. In general, the background PL can be calculated numerically by solving equations (C.1) and (C.3) on a 2D grid.

C.1. Equivalent symmetric problem

In experimental and numerical studies it is found that the incoherent steady-state polariton population $\tilde{N}_{ik}^{PL}$ has radial symmetry even under excitation with a specific wave vector [15, 37]. This property can be seen as peculiar of the quasi-elastic nature of the phonon-induced scattering $W_{iklk}^{(ph)}$ in this system, which is able to redistribute in a very short time the peaked coherent population in a symmetric pattern. Then the steady-state incoherent population depends only on the total flux of injected particles balanced by loss and no more on the specific form of $N_{ik}^{PL} \bigg|_{coh}$ at earlier times.

In our numerical studies this allows us to approximate the full 2D problem by an equivalent one with radial symmetry from the outset. The population calculated in this way should well reproduce the full 2D incoherent population once the steady-state is reached. Moreover, the symmetry of this equivalent system reduces the computation to the sole radial distribution for the population $N_{ik}^{PL}$, where $k = |k|$.

To do so we consider a radially symmetric pump

$$I_{ik} = I_0 e^{-\frac{(k-k_p)^2}{2\sigma^2}}, \quad (C.4)$$

which is chosen such that the flux of injected particles into the system is the same as in the case of a single pumped wave vector,
\[ \sum I_k = \sum I_k. \quad \text{(C.5)} \]

We change to polar coordinates,
\[ \sum k_{i,k} \simeq \frac{S}{(2\pi)^2} \int dk_x dk_y = \frac{S}{(2\pi)^2} \int k dk d\theta \simeq \sum k_{i,k} \left( \frac{S}{(2\pi)^2} \Delta k \Delta \theta \right), \quad \text{(C.6)} \]

and by making use of the fact that the distribution of populations is radially symmetric, \( N_{i\theta}^{PL} \equiv N_{i\theta}^{PL} \), we can manipulate the last term in equation (C.1) to obtain
\[ \partial t N_{i,k}^{PL} = -I_{i,k} N_{i,k}^{PL} + \bar{I}_{i,k} + \sum_{k'} W_{(i,k)(i',k')} N_{i',k'}^{PL}, \quad \text{(C.7)} \]

where we introduced \( W_{k,k'} = \sum_{\theta} W_{(i\theta),(i'\theta)} \). In figure 2 we solve the stationary solution of this equation and plot the stationary values of the incoherent part \( \bar{N}_{i,k}^{PL} \).

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