# Supporting Information

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# Coherence and Interaction in Confined Room-temperature Polariton Condensates with Frenkel Excitons

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# S1 Extension: Theoretical description of the blueshift in organic semiconductors

To describe microscopically the light-matter coupling in the system, we start with the description of a Frenkel exciton as a tightly bound electron-hole pair localized at an atomic site.[1] The electrons and holes are described by fermionic annihilation operators $\hat{a}_n$ and $\hat{b}_n$, respectively, and obey anticommutation relations: $\{\hat{a}_{n'}, \hat{a}_n^\dagger\} = \{\hat{b}_{n'}, \hat{b}_n^\dagger\} = \delta_{n',n}$ and $\{\hat{a}_{n'}, \hat{a}_n\} = \{\hat{b}_{n'}, \hat{b}_n\} = \{\hat{a}_{n'}, \hat{b}_n\} = 0$ for any $n, n'$. Here, the operator $\hat{a}_n^\dagger$ creates an electron in the excited state of an atom and the hole operator $\hat{b}_n^\dagger$ destroys the electron in the ground state (i.e. creates a hole). The site index $n$ runs from one to $N_s$, with the latter being the total number of available sites. The operator corresponding to the creation of the Frenkel exciton $n$ then reads $\hat{X}_n^\dagger = \hat{a}_n^\dagger \hat{b}_n^\dagger$ and acting on vacuum it generates a bound pair at position $n$, $|n\rangle = \hat{a}_n^\dagger \hat{b}_n^\dagger |\varnothing\rangle = \hat{X}_n^\dagger |\varnothing\rangle$. The binding energy of the Frenkel exciton comes from electron-hole onsite Coulomb interaction and is typically large ($> 100\,\mathrm{meV}$). The intersite Coulomb processes lead to delocalization of a pair and causes exciton-exciton interaction. Note that contrary to Wannier type excitons the conventional Coulomb exchange interaction between different sites is suppressed due to nearly zero overlap, as the system corresponds to the tight-binding limit. However, the intersite direct terms remain.

The system Hamiltonian can be written as a sum of free energies for the cavity mode $\hat{\mathcal{H}}_{\mathrm{cav}}$ and Frenkel excitons $\hat{\mathcal{H}}_X$, which are strongly coupled due to light-matter interaction. It reads

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_{\mathrm{cav}} + \hat{\mathcal{H}}_X + \hat{\mathcal{H}}_{\mathrm{coupl}} = \sum_{\mathbf{k}} \hbar\omega_{c,\mathbf{k}} \hat{c}_{\mathbf{k}}^\dagger \hat{c}_{\mathbf{k}} + \sum_n \Delta_n \hat{X}_n^\dagger \hat{X}_n + \sum_n G_{n,\mathbf{k}} (\hat{X}_n^\dagger \hat{c}_{\mathbf{k}} + h.c.), \qquad \text{(S1)}$$

where the cavity mode has the dispersion $\omega_{c,\mathbf{k}}$ and with $\hat{c}_{\mathbf{k}}$ ($\hat{c}_{\mathbf{k}}^\dagger$) being bosonic annihilation (creation) operators for a photon at momentum $\mathbf{k}$. The free energy of excitons $\hat{\mathcal{H}}_X$ [second term in Eq. (S1)] is described as a sum of two-level systems with transition energies $\Delta_n$.

Finally, the third term $\hat{\mathcal{H}}_{\text{coupl}}$ describing the light-matter coupling provides hybridization for the modes, where $G_{n,\mathbf{k}}$ is a coupling constant which in general can depend on the location of an atom and the wavevector of the cavity field.

To describe the coupling of the cavity photon to Frenkel excitons in the momentum space, the latter can be written as a delocalized mode using Fourier transform. The excitonic state reads

$$|X_{\mathbf{k}}\rangle = \hat{X}^{\dagger}_{\mathbf{k}}|\o\rangle = \frac{1}{\sqrt{N_s}}\sum_{n=1}^{N_s}\exp(i\mathbf{k}\cdot\mathbf{r}_n)|n\rangle, \tag{S2}$$

where $\mathbf{r}_n$ denotes positions of the localized excitations and $\mathbf{k}$ is an exciton momentum. Using the momentum space description, $\hat{\mathcal{H}}_{\text{coupl}}$ can be rewritten as

$$\hat{\mathcal{H}}_{\text{coupl}} = \sum_{\mathbf{k}} g_{\mathbf{k}}(\hat{X}^{\dagger}_{\mathbf{k}}\hat{c}_{\mathbf{k}} + h.c.), \tag{S3}$$

where we have redefined the coupling constant $g_{\mathbf{k}}$ to be dependent on the photon momentum, where the cavity photon is converted into an exciton with the same wavevector.

To describe the behavior of the system, it is instructive to write the Heisenberg equation of motion for the operators. Taking the Frenkel exciton mode with momentum $\mathbf{k}'$, we obtain

$$i\hbar\frac{\partial\hat{X}_{\mathbf{k}'}}{\partial t} = \left[\hat{X}_{\mathbf{k}'}, \hat{\mathcal{H}}_{\text{coupl}}\right] = \sum_{\mathbf{k}} g_{\mathbf{k}}\hat{c}_{\mathbf{k}}\left[\hat{X}_{\mathbf{k}'}, \hat{X}^{\dagger}_{\mathbf{k}}\right]. \tag{S4}$$

The central object in Eq. (S4) is the commutator for a Frenkel exciton mode. For excitons treated as ideal bosons, the commutator equals to the delta function and only $g_{\mathbf{k}'}$ remains in the $\mathbf{k}$-sum. This corresponds to the linear light-matter coupling term (Rabi splitting), which is usually considered in the low excitation density limit. In the case of increased carrier populations the composite nature of excitons, being formed by two fermions, modifies their behavior from being purely bosonic.[2] Given the specific structure of the Frenkel exciton, its statistics was separately studied.[3] For the localized exciton it can be derived straightforwardly from the electron-hole definition for $\hat{X}_n$ and reads

$$\left[\hat{X}_{n'}, \hat{X}_n^{\dagger}\right] = \delta_{n',n} - \hat{D}_{n',n} \equiv \delta_{n',n} - \delta_{n',n}(\hat{a}_n^{\dagger}\hat{a}_n + \hat{b}_n^{\dagger}\hat{b}_n). \tag{S5}$$

The last term in Eq. (S5) corresponds to the deviation from commutation relations and physically denotes the impossibility of double excitation at the single site. Importantly, the presence of the density-dependent term $\hat{D}_{n',n}$ makes the light-matter coupling effectively nonlinear.

To understand how the Rabi coupling effectively changes as a function of the exciton number (related to the pump intensity), we calculate the average of Eq. (S4) for the state which contains a certain number of Frenkel excitons. This state can be represented by a general density matrix $\rho^X$, which accounts for both thermal and coherent components. The average reads

$$\begin{aligned} i\hbar\langle\dot{\hat{X}}_{\mathbf{k}'}\rangle = & \quad \mathrm{Tr}\{\rho^X \sum_{\mathbf{k}} g_{\mathbf{k}}\langle\hat{c}_{\mathbf{k}}\rangle \left[\hat{X}_{\mathbf{k}'}, \hat{X}_{\mathbf{k}}^{\dagger}\right]\} = \\ = & \quad \sum_{\mathbf{k}} g_{\mathbf{k}}\langle\hat{c}_{\mathbf{k}}\rangle \sum_{lm} \rho_{lm}^X \langle N_m| \left[\hat{X}_{\mathbf{k}'}, \hat{X}_{\mathbf{k}}^{\dagger}\right] |N_l\rangle, \end{aligned} \tag{S6}$$

where we expanded the density matrix in the basis of many-body excitonic states $|N_l\rangle$, with the index $l$ denoting a certain distribution over the momentum index. Here, $\rho_{lm}^X$ corresponds to the occupations and coherences of the density matrix. The many-body states explicitly read $|N_l\rangle = \hat{X}_{\mathbf{k}_1}^{\dagger n_1^l}\hat{X}_{\mathbf{k}_2}^{\dagger n_2^l}..\hat{X}_{\mathbf{k}_S}^{\dagger n_S^l}|\text{ø}\rangle/\mathcal{N}$. $\mathcal{N}^2 := \langle N_l|N_l\rangle$ corresponds to the normalization constant and $n_j^l$ are numbers of excitons in the momentum mode $\mathbf{k}_j$.

First, in order to understand the influence of deviation from bosonic commutation relations, we perform averaging over generic many-body states and consider linear order corrections in the exciton density. We use the modified commutation relations for Frenkel excitons in the momentum space. The commutator generally has the same form $\left[\hat{X}_{\mathbf{k}'}, \hat{X}_{\mathbf{k}}^{\dagger}\right] = \delta_{\mathbf{k}',\mathbf{k}} - \hat{D}_{\mathbf{k}',\mathbf{k}}$, where the deviation operator can be conveniently defined through its action on the creation operators as[3]

$$\left[\hat{D}_{\mathbf{k}',\mathbf{k}}, \hat{X}^{\dagger}_{\mathbf{p}}\right] = \frac{2}{N_s}\hat{X}^{\dagger}_{\mathbf{p}+\mathbf{k}-\mathbf{k}'}. \tag{S7}$$

The latter can be seen as a consequence of nonzero portion of excitations as compared to the total number of states. Similarly, taking the commutator for creation of $N$ excitons, the commutator can be derived recursively as

$$\left[\hat{D}_{\mathbf{k}',\mathbf{k}}, (\hat{X}^{\dagger}_{\mathbf{p}})^{N}\right] = \frac{2N}{N_s}\hat{X}^{\dagger}_{\mathbf{p}+\mathbf{k}-\mathbf{k}'}(\hat{X}^{\dagger}_{\mathbf{p}})^{N-1}. \tag{S8}$$

The equation of motion for the exciton operator Eq. (S6) taken in a state $\rho^X$ can separated into diagonal and off-diagonal parts as

$$i\hbar\langle\dot{\hat{X}}_{\mathbf{k}'}\rangle = \sum_{\mathbf{k}} g_{\mathbf{k}}\langle\hat{c}_{\mathbf{k}}\rangle \sum_{l}\rho^{X}_{ll}\langle N_l|\left[\hat{X}_{\mathbf{k}'}, \hat{X}^{\dagger}_{\mathbf{k}}\right]|N_l\rangle + \sum_{\mathbf{k}} g_{\mathbf{k}}\langle\hat{c}_{\mathbf{k}}\rangle \sum_{l\neq m}\rho^{X}_{lm}\langle N_m|\left[\hat{X}_{\mathbf{k}'}, \hat{X}^{\dagger}_{\mathbf{k}}\right]|N_l\rangle, \tag{S9}$$

and we consider the two terms separately. The diagonal part is responsible for the leading processes which involve occupations and are present both before and after condensation. The off-diagonal terms are only present after condensation, where coherence $\rho^X_{lm}$ $(l \neq m)$ is developed.

The diagonal part of the commutator reads

$$\begin{aligned}
&\sum_{\mathbf{k}} g_{\mathbf{k}}\langle\hat{c}_{\mathbf{k}}\rangle \sum_{l}\rho^{X}_{ll}\langle N_l|\left[\hat{X}_{\mathbf{k}'}, \hat{X}^{\dagger}_{\mathbf{k}}\right]|N_l\rangle = g_{\mathbf{k}'}\langle\hat{c}_{\mathbf{k}'}\rangle - \\
&-\sum_{\mathbf{k}} g_{\mathbf{k}}\langle\hat{c}_{\mathbf{k}}\rangle \sum_{l}\rho^{X}_{ll}\frac{1}{\langle N_l|N_l\rangle}\Big(\langle\emptyset|\hat{X}^{n^l_S}_{\mathbf{k}_S}..\hat{X}^{n^l_2}_{\mathbf{k}_2}\hat{X}^{n^l_1}_{\mathbf{k}_1}\left[\hat{D}_{\mathbf{k}',\mathbf{k}}, \hat{X}^{\dagger n^l_1}_{\mathbf{k}_1}\right]\hat{X}^{\dagger n^l_2}_{\mathbf{k}_2}..\hat{X}^{\dagger n^l_S}_{\mathbf{k}_S}|\emptyset\rangle \\
&+\langle\emptyset|\hat{X}^{n^l_S}_{\mathbf{k}_S}..\hat{X}^{n^l_2}_{\mathbf{k}_2}\hat{X}^{n^l_1}_{\mathbf{k}_1}\hat{X}^{\dagger n^l_1}_{\mathbf{k}_1}\left[\hat{D}_{\mathbf{k}',\mathbf{k}}, \hat{X}^{\dagger n^l_2}_{\mathbf{k}_2}\right]..\hat{X}^{\dagger n^l_S}_{\mathbf{k}_S}|\emptyset\rangle + ...\Big),
\end{aligned} \tag{S10}$$

where the first term in the RHS $g_{\mathbf{k}'}\langle\hat{c}_{\mathbf{k}'}\rangle$ corresponds to the coupling in the weak excitation limit. It directly results into a lowest order light-matter coupling term of $g_0 \equiv g$, where we consider only the modes coupled to light, and set $k' = 0$. The second term then corresponds to the deviation of statistics for the excited system. Its parts depend on the commutation of the deviation operator with different momentum states (taken to be $\{\mathbf{k}_1, \mathbf{k}_2, ..., \mathbf{k}_S\}$), weighted with a probability distribution. Looking into the first commutator as an example, we see that

$$\sum_{\mathbf{k}} g_{\mathbf{k}} \langle \hat{c}_{\mathbf{k}} \rangle \sum_l \rho_{ll}^X \frac{1}{\langle N_l | N_l \rangle} \langle \o | \hat{X}_{\mathbf{k}_S}^{n_S^l} .. \hat{X}_{\mathbf{k}_2}^{n_2^l} \hat{X}_{\mathbf{k}_1}^{n_1^l} \left[ \hat{D}_{\mathbf{k}',\mathbf{k}}, \hat{X}_{\mathbf{k}_1}^{\dagger n_1^l} \right] \hat{X}_{\mathbf{k}_2}^{\dagger n_2^l} .. \hat{X}_{\mathbf{k}_S}^{\dagger n_S^l} | \o \rangle = \tag{S11}$$

$$= \sum_{\mathbf{k}} g_{\mathbf{k}} \langle \hat{c}_{\mathbf{k}} \rangle \sum_l \rho_{ll}^X \frac{1}{\langle N_l | N_l \rangle} \frac{2n_1^l}{N_s} \langle \o | \hat{X}_{\mathbf{k}_S}^{n_S^l} .. \hat{X}_{\mathbf{k}_2}^{n_2^l} \hat{X}_{\mathbf{k}_1}^{n_1^l} \hat{X}_{\mathbf{k}_1+\mathbf{k}-\mathbf{k}'}^{\dagger} \hat{X}_{\mathbf{k}_1}^{\dagger (n_1^l - 1)} \hat{X}_{\mathbf{k}_2}^{\dagger n_2^l} .. \hat{X}_{\mathbf{k}_S}^{\dagger n_S^l} | \o \rangle \approx$$

$$\approx \frac{2n_1^l}{N_s} g_{\mathbf{k}'} \langle \hat{c}_{\mathbf{k}'} \rangle \sum_l \rho_{ll}^X ,$$

where in the last equality we considered the dominant contribution which appears for $\mathbf{k} = \mathbf{k}'$, the ket-state reduces to the original $|N_l\rangle$ and we further note that $\sum_l \rho_{ll}^X = 1$ due to normalization. While other contributions may appear for $\mathbf{k} \neq \mathbf{k}'$, this shall happen in higher orders of $N_s^{-1}$. Finally, performing the same commutation for different momentum modes, we can write the diagonal contribution as

$$i\hbar \langle \dot{\hat{X}}_{\mathbf{k}'} \rangle = \sum_{\mathbf{k}} g_{\mathbf{k}} \langle \hat{c}_{\mathbf{k}} \rangle \sum_l \rho_{ll}^X \langle N_l | \left[ \hat{X}_{\mathbf{k}'}, \hat{X}_{\mathbf{k}}^{\dagger} \right] | N_l \rangle \approx \tag{S12}$$

$$\approx g_{\mathbf{k}'} \langle \hat{c}_{\mathbf{k}'} \rangle - 2 g_{\mathbf{k}'} \langle \hat{c}_{\mathbf{k}'} \rangle \sum_l \rho_{ll}^X \frac{(n_1^l + n_2^l + .. + n_S^l)}{N_s} = g_{\mathbf{k}'} \langle \hat{c}_{\mathbf{k}'} \rangle \Big( 1 - \frac{2N_{\mathrm{tot}}^X}{N_s} \Big),$$

where $N_{\mathrm{tot}}^X$ corresponds to the total number of Frenkel excitons, summed over the distribution in the momentum space. The off-diagonal terms can be written in the same way. However, as they are proportional to coherences, this term becomes non-zero only after the threshold and we generally can neglect it in the lowest order. By deriving Eq. (S12) we draw an important conclusion: the presence of excitons always leads to space phase filling, no matter the way they are distributed.

In Eq. (S12) we accounted for the linear term in $N_s^{-1}$ only, which corresponds to the lowest density-dependent correction to the light-matter coupling, and so far only excitonic EOM renormalization was considered. It is important to note that since we assume all excitons to couple to the cavity mode in the Hamiltonian (S3), the exciton number $N^X_{\text{tot}} = N^X_{\text{PL}}$ corresponds only to excitons coupled to light. Eq. (S12) thus describes the decrease of the light-matter coupling for the increased total population of Frenkel excitons, as long as they lie within the light cone. The exciton density dependence of the light-matter coupling thus introduces the optically nonlinear behavior, which was observed experimentally.

Next, we consider the influence of higher-order corrections for the light-matter interaction term. To extract the renormalized Rabi frequency with nonlinear corrections, we need to derive the dispersion relations for lower and upper polariton modes accounting for the phase space filling. To do so, we consider the Hamiltonian (S1) averaged over a many-body state with excitons occupying $\mathbf{k} \approx 0$ mode, being a reasonable approximation for the effectively 0D dimple cavity. Then, following the usual strategy the dispersions are inferred from its diagonalization in "photons"-"excitons" basis, where light-matter coupling represents a cross term.[4] The Hamiltonian energy for a small wavevector reads

$$\langle \hat{\mathcal{H}}_{\mathbf{k}} \rangle = \hbar\omega_c \langle \hat{c}^\dagger \hat{c} \rangle + \Delta \langle \hat{X}^\dagger_{\mathbf{k}} \hat{X}_{\mathbf{k}} \rangle + g(0)(\langle \hat{X}^\dagger_{\mathbf{k}} \hat{c} \rangle + \langle \hat{c}^\dagger \hat{X}_{\mathbf{k}} \rangle), \tag{S13}$$

where we also set the value of light-matter coupling to the coupling constant taken at the small photon momentum $g(0)$, which is superradiantly enhanced. The averages for $\langle \hat{c}^\dagger \hat{c} \rangle$ and $\langle \hat{X}^\dagger_{\mathbf{k}} \hat{X}_{\mathbf{k}} \rangle$ terms provide photon and exciton numbers $N_c, N_X$ multiplied by corresponding energies. For the coupling terms, if Frenkel excitons corresponded to ideal bosons, the averages for $\langle \hat{X}^\dagger_{\mathbf{k}} \hat{c} \rangle$ and $\langle \hat{c}^\dagger \hat{X}_{\mathbf{k}} \rangle$ terms would provide a prefactor of $\sqrt{N_c N_X}$. Finally, normalizing the energy per single polariton, the dispersions can be obtained by simple diagonalization of the two-by-two matrix

$$\begin{pmatrix} \hbar\omega_c & g(0) \\ g(0) & \Delta \end{pmatrix} \Rightarrow \hbar\omega_{L,U} = \frac{\hbar\omega_c + \Delta}{2} \mp \frac{1}{2}\sqrt{(\hbar\omega_c - \Delta)^2 + 4g(0)^2}, \tag{S14}$$

where $\hbar\omega_{L,U}$ are lower and upper polariton energies, respectively. Now, we need to account for the phase space filling corrections which modify the off-diagonal terms. The central object here corresponds to the expectation value of $\langle \hat{c}^\dagger \hat{X}_{\mathbf{k}} \rangle$, which is derived considering transition from $|N_c - 1, N_X\rangle$ to $|N_c, N_X - 1\rangle$ state. The expectations for photons and excitons can be calculated separately, where the first one trivially reads $\langle N_c|\hat{c}^\dagger|N_c - 1\rangle = \sqrt{N_c}$. The second contribution comes from the excitonic expectation performed for states $|N_X\rangle = \hat{X}_0^{\dagger N_X}|\text{ø}\rangle/\mathcal{N}_{N_X}$ and $|N_X - 1\rangle = \hat{X}_0^{\dagger(N_X-1)}|\text{ø}\rangle/\mathcal{N}_{N_X-1}$, where $\mathcal{N}_{N_X} = \sqrt{\langle \text{ø}|\hat{X}_0^{N_X}\hat{X}_0^{\dagger N_X}|\text{ø}\rangle}$, $\mathcal{N}_{N_X-1}$ correspond to normalization prefactors for each state. The matrix element

$$\langle N_X - 1|\, \hat{X}_{\mathbf{k}}\, |N_X\rangle = \frac{1}{\mathcal{N}_{N_X}\mathcal{N}_{N_X-1}} \langle \text{ø}|\hat{X}_0^{N_X-1}[\hat{X}_{\mathbf{k}}, \hat{X}_0^{\dagger N_X}]|\text{ø}\rangle \tag{S15}$$

can be further rewritten using known commutation rules for Frenkel excitons[3]

$$[\hat{X}_{\mathbf{q}}, \hat{X}_{\mathbf{q}'}^{\dagger N_X}] = N_X(\hat{X}_{\mathbf{q}'}^\dagger)^{N_X-1}(\delta_{\mathbf{q},\mathbf{q}'} - \hat{D}_{\mathbf{q},\mathbf{q}'}) - \frac{N_X(N_X-1)}{N_s}\hat{X}_{2\mathbf{q}'-\mathbf{q}}^\dagger(\hat{X}_{\mathbf{q}'}^\dagger)^{N_X-2} \tag{S16}$$

together with Eq. (S8), as well as closure relations $\mathcal{N}_{N_X-n}^2 = \langle \text{ø}|\hat{X}_0^{N_X-n}\hat{X}_0^{\dagger N_X-n}|\text{ø}\rangle = (N_X - n)!F_{N_X-n}$, where

$$F_{N_X-n} = \frac{(N_s - N_X)!N_s^n}{(N_s - N_X + n)!}F_{N_X}. \tag{S17}$$

Then, Eq. (S15) can be rewritten recursively as

$$\langle N_X - 1| \hat{X}_{\mathbf{k}} |N_X\rangle = \sqrt{N_X}\sqrt{\frac{(N_s - N_X + 1)}{N_s}}\Big[\frac{F_{N_X-1}}{F_{N_X}}\delta_{0,\mathbf{k}} - (N_X - 1)\frac{F_{N_X-2}}{F_{N_X}}\delta_{0,\mathbf{k}} + (N_X - 1)(N_X - 2)\frac{F_{N_X-3}}{F_{N_X}}\delta_{0,\mathbf{k}} - ...\Big]. \tag{S18}$$

The closed expression can be derived when summed up to $N_X = N_s$, corresponding to fully saturated system. Finally, in the relevant limit of $N_s \ll 1$ and $N_X > 2$ we can write the matrix element as

$$\langle N_X - 1| \hat{X}_{\mathbf{k}} |N_X\rangle \approx \sqrt{N_X}\sqrt{1 - \frac{N_X}{N_s}}\delta_{0,\mathbf{k}}. \tag{S19}$$

This leads to effective renormalization of light-matter coupling as

$$g(N_X) = g(0)\sqrt{1 - \frac{N_X}{N_s}}. \tag{S20}$$

Finally, using the same approach as for ideal bosons, the dispersions can be derived using density-depended coupling

$$\begin{pmatrix} \hbar\omega_c & g(0)\sqrt{1 - N_X/N_s} \\ g(0)\sqrt{1 - N_X/N_s} & \Delta \end{pmatrix} \Rightarrow \hbar\omega_{L,U}(N_X). \tag{S21}$$

## S2 Nonlinearity of the samples s1-s3 and excitation power-dependent analysis of the device s3

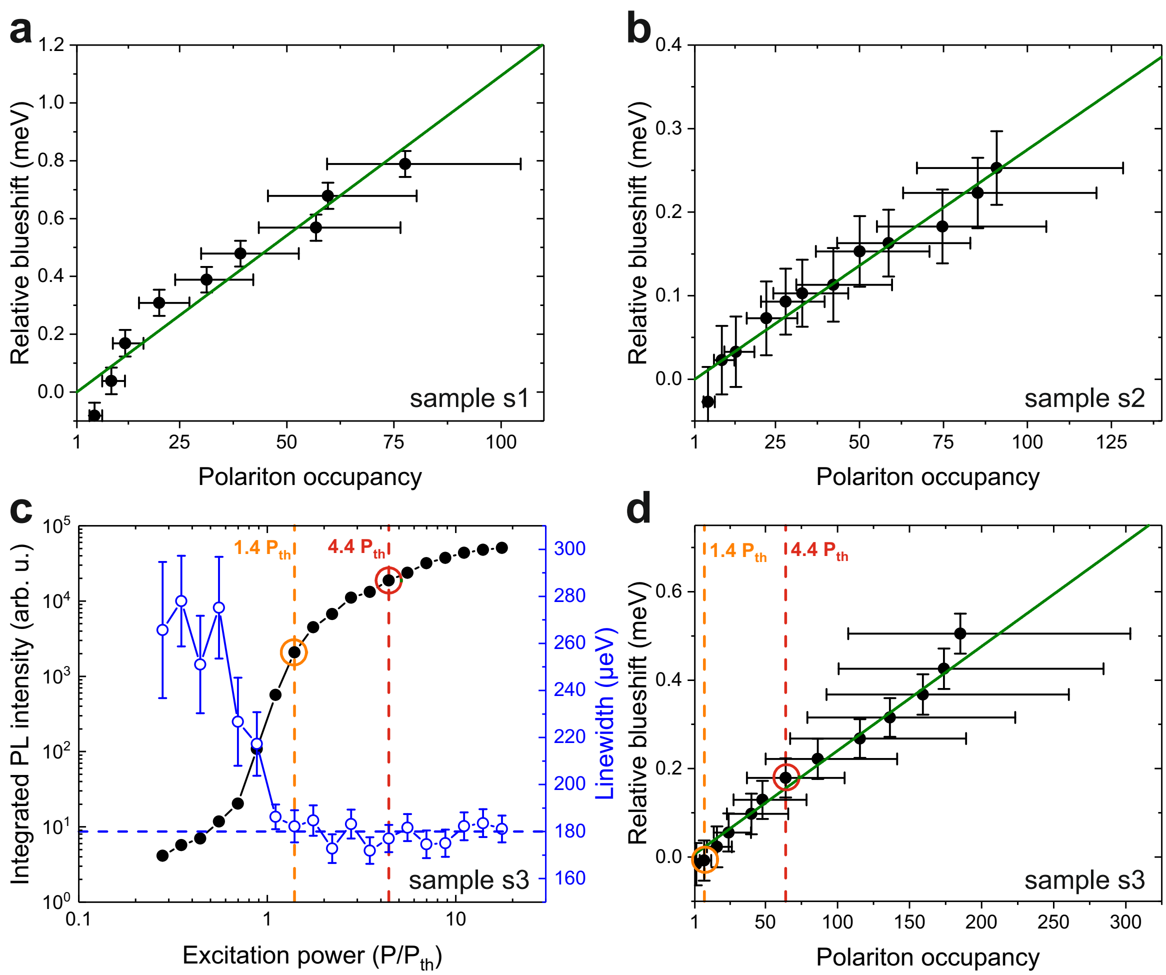


Figure S1: (a), (b), (d) Relative blueshift versus polariton occupancy of the condensate for the different samples s1-s3. A linear increase is clearly visible. The excitation powers used in Figure 5 in the main text are highlighted in (d). (c) Excitation power-dependent analysis of the device s3, which was used for the coherence measurements (Figure 5 in the main text). Integrated emission intensity (black filled circles) versus normalized excitation intensity. At the threshold power $P_{th}$ a significant increase in the intensity as well as a reduction of the linewidth below the resolution limit of the spectrometer (180 $\mu$eV, blue dashed line) is observed.

## S3 Temporal coherence of the polariton condensate

To describe the decay of $g^{(1)}(\tau)$ for different polariton occupancies $\tilde{n}$ we use an analytical model of single-mode matter-wave lasers,[5–7] which takes single particle decay processes, gain, gain saturation as well as effective interactions within the lasing mode simultaneously into account. The $g^{(1)}(\tau)$ is then given by[8]

$$\left|g^{(1)}(\tau)\right| = \exp\left(-\frac{4(n_s+\tilde{n})u^2}{\tilde{\gamma}^2}\left(\exp\left(-\tilde{\gamma}\tau\right)+\tilde{\gamma}-1\right)\right)\times\exp\left(\frac{n_s+\tilde{n}}{4\tilde{n}^2}\left(\exp\left(-\tilde{\gamma}\tau\right)-\tilde{\gamma}-1\right)\right), \tag{S22}$$

where $\tilde{\gamma} = \frac{\tilde{n}}{n_s+\tilde{n}}\gamma$, $\gamma$ is the decay rate of the polariton ground state, $n_s$ is the gain saturation number (which corresponds to the depletion of the pump reservoir by processes which populate the condensate[7]) and $u$ is an effective interaction constant. Using Eq. (S22), the decay of $g^{(1)}(\tau)$ for different polariton occupancies (Fig. 5c-d of the main text) can simultaneously fitted with shared parameters for $\gamma$, $n_s$ and $u$ (see Fig. S2). Note, that the results for $\gamma = 0.085 \pm 0.014\,\mathrm{ps}^{-1}$ and $n_s = 7.3 \pm 1.5$ are very reasonable, and the result for $\hbar u V_c = (2.0 \pm 0.2)\,\mu\mathrm{eV}\times\mu\mathrm{m}^3$ per polariton in the condensate is in excellent agreement with the analysis provided by tracing the condensate energy with pump power, which yields a value of $\hbar u V_c = (2.7^{+2.5}_{-1.1})\,\mu\mathrm{eV}\times\mu\mathrm{m}^3$ per polariton in the condensate. Using those paramters, the temporal coherence $\tau_c$ of the polariton condensate for different mean occupancies $\tilde{n}$ can be calculates (see Fig. 5e of the main text).

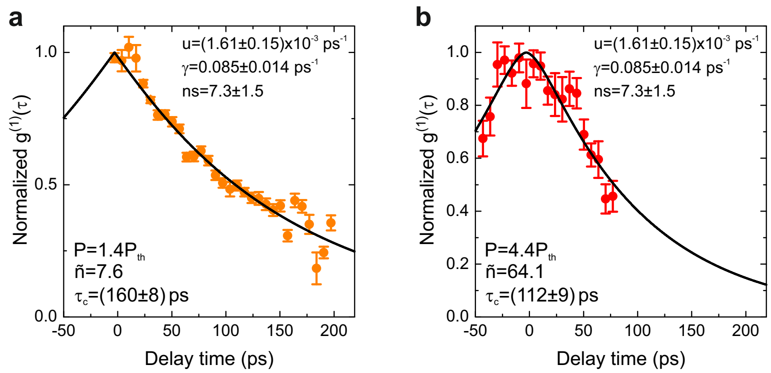


Figure S2: Normalized measured first-order coherence $g^{(1)}(\tau)$ for different delay times for a pump power of $P = 1.4P_{th}$ (a) and $P = 4.4P_{th}$ (b). The curves were fitted simultaneously using Eq. (S22) and shared parameters for $\gamma$, $n_s$ and $u$.

# References


(1) Combescot, M.; Pogosov, W. Microscopic derivation of Frenkel excitons in second quantization. *Phys. Rev. B* **2008**, *77*, 085206.

(2) Combescot, M.; Betbeder-Matibet, O.; Dubin, F. The many-body physics of composite bosons. *Phys. Rep.* **2008**, *463*, 215–320.

(3) Combescot, M.; Pogosov, W. Composite boson many-body theory for Frenkel excitons. *The European Physical Journal B* **2009**, *68*, 161–181.

(4) Kavokin, A. V.; Baumberg, J. J.; Malpuech, G.; Laussy, F. P. *Microcavities*, 2nd ed.; Series on Semiconductor Science and Technology; Oxford University Press, 2017.

(5) Wiseman, H. M. Defining the (atom) laser. *Phys. Rev. A* **1997**, *56*, 2068–2084.

(6) Thomsen, L. K.; Wiseman, H. M. Atom-laser coherence and its control via feedback. *Phys. Rev. A* **2002**, *65*, 063607.

(7) Whittaker, D. M.; Eastham, P. R. Coherence properties of the microcavity polariton condensate. *EPL (Europhysics Letters)* **2009**, *87*, 27002.

(8) Kim, S.; Zhang, B.; Wang, Z.; Fischer, J.; Brodbeck, S.; Kamp, M.; Schneider, C.; Höfling, S.; Deng, H. Coherent Polariton Laser. *Phys. Rev. X* **2016**, *6*, 011026.