

SUPPLEMENTARY INFORMATION:
**Indistinguishability and correlations of photons generated
by quantum emitters undergoing spectral diffusion**

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S1. THE DETECTOR SPECTROGRAM

The photon coincidence signal Eq. (7) is defined in terms of the time-and-frequency resolved electric field $E^{(tf)}$. A frequency (spectral) gate is combined with time gate. The detector with input located at r_D is represented by a time gate F_t centered at \bar{t} followed by a frequency gate F_f centered at $\bar{\omega}$ [1]. First, the time gate transforms the electric field at a point in the sample r_S : $E(r_S, t) = \sum_q \hat{E}_q(r_S, t)$ with $\hat{E}_q(r_S, t) = E(r_S, \omega_q) e^{-i\omega_q t}$ as follows:

$$E^{(t)}(\bar{t}; r_S, t) = F_t(t, \bar{t}) E(r_S, t). \quad (\text{S1})$$

Then, the frequency gate is applied $E^{(tf)}(\bar{t}, \bar{\omega}; r_S, \omega) = F_f(\omega, \bar{\omega}) E^{(t)}(\bar{t}; r_S, \omega)$ to obtain the time-and-frequency-gated field. The combined detected field at r_D can be written as

$$E^{(tf)}(\bar{t}, \bar{\omega}; r_D, t) = \int_{-\infty}^{\infty} dt' F_f(t - t', \bar{\omega}) F_t(t', \bar{t}) E(r_S, t'), \quad (\text{S2})$$

where $E(t) \equiv \sum_s \sqrt{2\pi\hbar\omega_s/\Omega\hat{a}_s} e^{-\omega_s t}$ and Ω is a mode quantization volume. For clarity we hereafter omit the position dependence in the fields and include the propagation between r_S and r_D in the spectral gate function. Using Eq. (7) - (9) we next define the detector spectrogram for the j -th detector, $j = r, s$

$$W_D^{(j)}(\bar{t}_j, \bar{\omega}_j; t', \omega'; T) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} |F_f^{(j)}(\omega, \bar{\omega}_j)|^2 W_t^{(j)}(t', \omega' - \omega, \bar{t}_j) e^{-i\omega T}, \quad (\text{S3})$$

where

$$W_t^{(j)}(t', \omega) = \int_{-\infty}^{\infty} d\tau F_t^{(j)*}(t' + \tau, \bar{t}) F_t^{(j)}(t', \bar{t}) e^{i\omega\tau}. \quad (\text{S4})$$

We can freely vary the parameters of $F_f^{(j)}$ and $F_t^{(j)}$. However the temporal $\tilde{\sigma}_T^j$ and spectral $\tilde{\sigma}_\omega^j$ resolutions of the spectrogram (S3) will always satisfy the Fourier uncertainty $\tilde{\sigma}_\omega^j / \tilde{\sigma}_T^j > 1$. Assuming that time and frequency gates are Gaussian

$$F_t^{(j)}(t, \bar{t}_j) = e^{-\frac{1}{2}\sigma_T^{j2}(t-\bar{t}_j)^2}, \quad F_f^{(j)}(\omega, \bar{\omega}_j) = e^{-\frac{(\omega-\bar{\omega}_j)^2}{4\sigma_\omega^{j2}}}, \quad (\text{S5})$$

we obtain the Wigner spectrogram of the detector

$$W_D^{(j)}(\bar{t}_j, \bar{\omega}_j; t', \omega'; T) = \frac{1}{\sigma_T^j [(\sigma_\omega^j)^{-2} + (\sigma_T^j)^{-2}]^{1/2}} e^{-\frac{1}{2}\tilde{\sigma}_T^{j2}(t'-\bar{t}_j)^2 - \frac{(\omega'-\bar{\omega}_j)^2}{2\tilde{\sigma}_\omega^{j2}} - iA_j(\omega'-\bar{\omega}_j)(t'-\bar{t}_j + C_j T) - \frac{1}{2}q_j^2 T^2 - i\bar{\omega}_j T}, \quad (\text{S6})$$

where $\tilde{\sigma}_T^{j2} = \sigma_T^{j2} + [(\sigma_T^j)^{-2} + (\sigma_\omega^j)^{-2}]^{-1}$, $\tilde{\sigma}_\omega^{j2} = \sigma_T^{j2} + \sigma_\omega^{j2}$, $A_j = \sigma_T^{j2}[\sigma_T^{j2} + \sigma_\omega^{j2}]^{-1}$, $C_j = \sigma_\omega^{j2}/\sigma_T^{j2}$, and $q_j^{-2} = (\sigma_T^j)^{-2} + (\sigma_\omega^j)^{-2}$. Using Eq. (S3) one can recast the signal (7) in the form of Eq. (9), where the bare spectrogram is given by Eqs. (S12) - (S13).

S2. SPECTRAL DIFFUSION

We assume that the electronic states of molecule $\alpha = a, b$ are coupled to a harmonic bath described by the Hamiltonian $\hat{H}_B^\alpha = \sum_k \hbar\omega_k (\hat{a}_k^\dagger \hat{a}_k^\alpha + 1/2)$. The bath perturbs the energy of state ν . This is represented by the Hamiltonian

$$\hat{H}_\nu^\alpha = \hbar^{-1} \langle \nu_\alpha | \hat{H} | \nu_\alpha \rangle = \epsilon_{\nu_\alpha} + \hat{q}_{\nu_\alpha} + \hat{H}_B^\alpha, \quad (\text{S7})$$

where \hat{q}_ν is a collective bath coordinate

$$\hat{q}_{\nu_\alpha} = \hbar^{-1} \langle \nu_\alpha | \hat{H}_{SB} | \nu_\alpha \rangle = \sum_k d_{\nu_\alpha \nu_\alpha, k} (\hat{a}_k^\dagger + \hat{a}_k), \quad (\text{S8})$$

$d_{mn,k}$ represents bath-induced fluctuations of the transition energies ($m = n$) and the intermolecular coupling ($m \neq n$). We define the line-shape function

$$g_\alpha(t) \equiv g_{\nu_\alpha \nu'_\alpha}(t) = \int \frac{d\omega}{2\pi} \frac{C''_{\nu_\alpha \nu'_\alpha}(\omega)}{\omega^2} \left[\coth\left(\frac{\beta\hbar\omega}{2}\right) (1 - \cos\omega t) + i \sin\omega t - i\omega t \right], \quad (\text{S9})$$

where the bath spectral density is given by

$$C''_{\nu_\alpha\nu'_\alpha}(\omega) = \frac{1}{2} \int_0^\infty dt e^{i\omega t} \langle [\hat{q}_{\nu_\alpha}(t), \hat{q}_{\nu'_\alpha}(0)] \rangle, \quad (\text{S10})$$

$\beta = k_B T$ with k_B being the Boltzmann constant and T is the ambient temperature. The matter correlation function can be evaluated using the second order cumulant expansion using Eq. (10). We shall use the overdamped Brownian oscillator model for the spectral density, assuming a single nuclear coordinate ($\nu_\alpha = \nu'_\alpha$)

$$C''_{\nu_\alpha\nu_\alpha}(\omega) = 2\lambda_\alpha \frac{\omega\Lambda_\alpha}{\omega^2 + \Lambda_\alpha^2}, \quad (\text{S11})$$

where Λ_α is the fluctuation relaxation rate and λ_α is the system-bath coupling strength. The corresponding lineshape function $g_\alpha(t)$ in the high temperature limit $k_B T \gg \hbar\Lambda_\alpha$ is then given by Eq. (11).

S3. THE COINCIDENCE SIGNAL

The signal Eq. (7) is guaranteed to be positive since it can be recast as a modulus square of an amplitude. By treating explicitly the detector spectrogram (S3) we now define a “bare signal” - a quantity that contains the information about the field-matter interactions only and is independent of the detection. The bare signal is not an observable. The loop diagrams which represent the process of excitation by incoming pulse and spontaneous emission of the photon are depicted in Fig. 1c. In order to maintain the bookkeeping for all interactions and develop a perturbative expansion for signals we describe the signal in terms of Liouville space “left” and “right” superoperators. With each ordinary operator A we can associate a pair of superoperators [2] $\hat{A}_L X = AX$, $\hat{A}_R X = XA$, and $\hat{A}_- = \hat{A}_L - \hat{A}_R$. To avoid confusion and distinguish the ordinary operators (e.g. A) from the superoperator quantities we hereafter denote all superoperators by “hat” (e.g. \hat{A}). By taking into account the input-output transformation of a beam splitter in Eq. (8) and relation (9), the Wigner spectrograms of the bare signal $R_B^{(i)}(t'_s, \omega'_s; t'_r, \omega'_r)$ and $R_B^{(ii)}(t'_s, \omega'_s; t'_r, \omega'_r)$ may be recast in terms of superoperators using the diagram shown in Fig. 1c:

$$R_B^{(i)}(t'_s, \omega'_s; t'_r, \omega'_r) = \sum_{u,u'} \sum_{v,v'} \int_{-\infty}^{\infty} d\tau_s d\tau_r e^{-i\omega'_s \tau_s - i\omega'_r \tau_r} \\ \times \langle \mathcal{T} \hat{E}_{u'R}^\dagger(t'_s + \tau_s, r_b) \hat{E}_{v'R}^\dagger(t'_r + \tau_r, r_a) \hat{E}_{vL}(t'_r, r_a) \hat{E}_{uL}(t'_s, r_b) e^{-\frac{i}{\hbar} \int_{-\infty}^{\infty} \hat{H}^L(T) dT} \rangle, \quad (\text{S12})$$

$$R_B^{(ii)}(t'_s, \omega'_s; t'_r, \omega'_r) = - \sum_{u,u'} \sum_{v,v'} \int_{-\infty}^{\infty} d\tau_s d\tau_r e^{-i\omega'_s \tau_s - i\omega'_r \tau_r} \\ \times \langle \mathcal{T} \hat{E}_{u'R}^\dagger(t'_s + \tau_s, r_b) \hat{E}_{v'R}^\dagger(t'_r + \tau_r, r_a) \hat{E}_{uL}(t'_s, r_a) \hat{E}_{vL}(t'_r, r_b) e^{-\frac{i}{\hbar} \int_{-\infty}^{\infty} \hat{H}^L(T) dT} \rangle, \quad (\text{S13})$$

where the angular brackets denote $\langle \dots \rangle \equiv \text{Tr}[\rho_0 \dots]$ with ρ_0 is initial density operator defined in the joint field-matter space of the entire system. The Hamiltonian superoperator is given by

$$\hat{H}'_\nu(t) = \hat{E}_\nu^\dagger(t) \hat{V}_\nu(t) + H.c., \quad \nu = L, R. \quad (\text{S14})$$

Our key bookkeeping device is the time ordering superoperator \mathcal{T}

$$\mathcal{T} \hat{E}_\nu(t_1) \hat{E}_{\nu'}(t_2) = \hat{E}_\nu(t_1) \hat{E}_{\nu'}(t_2) \theta(t_1 - t_2) + \hat{E}_{\nu'}(t_2) \hat{E}_\nu(t_1) \theta(t_2 - t_1), \quad (\text{S15})$$

where $\theta(t)$ is the Heaviside step function. Note that the electric field in the correlation function in Eq. (S12) is a product of contributions from molecules a and b whereas Eq. (S13) shows that the photon is generated by a pair of molecules according to the diagrams in Fig. 1c. We further note, that when working in the field space alone, the number of independent field modes become restricted to 2: $u = u'$ and $v = v'$. However, this is not the case in the field plus matter joint space. By expanding the exponential operator in Eqs. - (S12) - (S13) to fourth order for each molecule we obtain

$$R_B^{(i)}(t'_s, \omega'_s; t'_r, \omega'_r) \\ = \frac{1}{\hbar^8} \sum_{u,u'} \sum_{v,v'} \int_{-\infty}^{\infty} d\tau_s d\tau_r e^{-i\omega'_s \tau_s - i\omega'_r \tau_r} \int_{-\infty}^{t'_r} d\tau_1 \int_{-\infty}^{\tau_1} d\tau_2 \int_{-\infty}^{t'_r + \tau_r} d\tau'_1 \int_{-\infty}^{\tau'_1} d\tau'_2 \int_{-\infty}^{t'_s} d\tau_3 \int_{-\infty}^{\tau_3} d\tau_4 \int_{-\infty}^{t'_s + \tau_s} d\tau'_3 \int_{-\infty}^{\tau'_3} d\tau'_4 \\ \times \langle \mathcal{T} \hat{E}_{u'R}^\dagger(t'_s + \tau_s, r_b) \hat{E}_{v'R}^\dagger(t'_r + \tau_r, r_a) \hat{E}_{vL}(t'_r, r_a) \hat{E}_{uL}(t'_s, r_b) \hat{E}_{u'R}(\tau'_3, r_b) \hat{E}_{v'R}(\tau'_1, r_a) \hat{E}_{vL}^\dagger(\tau_1, r_a) \hat{E}_{uL}^\dagger(\tau_3, r_b) \rangle \\ \times \mathcal{E}_p^*(\tau'_2, r_a) \mathcal{E}_p(\tau_2, r_a) \mathcal{E}_p^*(\tau'_4, r_b) \mathcal{E}_p(\tau_4, r_b) \langle \langle gg | \mathcal{T} \hat{V}_R^\dagger(\tau'_1) \hat{V}_L(\tau_1) \hat{V}_R(\tau'_2) \hat{V}_L^\dagger(\tau_2) | gg \rangle \rangle_a \langle \langle gg | \mathcal{T} \hat{V}_R^\dagger(\tau'_3) \hat{V}_L(\tau_3) \hat{V}_R(\tau'_4) \hat{V}_L^\dagger(\tau_4) | gg \rangle \rangle_b, \quad (\text{S16})$$

$$\begin{aligned}
R_B^{(ii)}(t'_s, \omega'_s; t'_r, \omega'_r) &= -\frac{1}{\hbar^8} \sum_{u, u'} \sum_{v, v'} \int_{-\infty}^{\infty} d\tau_s d\tau_r e^{-i\omega'_s \tau_s - i\omega'_r \tau_r} \int_{-\infty}^{t'_s} d\tau_1 \int_{-\infty}^{\tau_1} d\tau_2 \int_{-\infty}^{t'_r + \tau_r} d\tau'_1 \int_{-\infty}^{\tau'_1} d\tau'_2 \int_{-\infty}^{t'_r} d\tau_3 \int_{-\infty}^{\tau_3} d\tau_4 \int_{-\infty}^{t'_s + \tau_s} d\tau'_3 \int_{-\infty}^{\tau'_3} d\tau'_4 \\
&\times \langle \mathcal{T} \hat{E}_{u'R}^\dagger(t'_s + \tau_s, r_b) \hat{E}_{v'R}^\dagger(t'_r + \tau_r, r_a) \hat{E}_{uL}(t'_s, r_a) \hat{E}_{vL}(t'_r, r_b) \hat{E}_{u'R}(\tau'_3, r_b) \hat{E}_{v'R}(\tau'_1, r_a) \hat{E}_{uL}(\tau_1, r_a) \hat{E}_{vL}(\tau_3, r_b) \rangle \\
&\times \mathcal{E}_p^*(\tau'_2, r_a) \mathcal{E}_p(\tau_2, r_a) \mathcal{E}_p^*(\tau'_4, r_b) \mathcal{E}_p(\tau_4, r_b) \langle \langle gg | \mathcal{T} \hat{V}_R^\dagger(\tau'_1) \hat{V}_L(\tau_1) \hat{V}_R(\tau'_2) \hat{V}_L^\dagger(\tau_2) | gg \rangle \rangle_a \langle \langle gg | \mathcal{T} \hat{V}_R^\dagger(\tau'_3) \hat{V}_L(\tau_3) \hat{V}_R(\tau'_4) \hat{V}_L^\dagger(\tau_4) | gg \rangle \rangle_b,
\end{aligned} \tag{S17}$$

where $\langle \langle gg | A | gg \rangle \rangle \equiv \text{Tr}[|g\rangle\langle A | g\rangle\langle g|]$ and we have replaced a classical excitation field by its expectation value. Since the spontaneous emission modes u, u', v, v' are initially in the vacuum state, we must expand to second order in each mode. It is clear that the process of coincidence counting involves four radiation modes, in contrast to the field space analysis [3]. We further evaluate the time integrals using the explicit time dependence of the spontaneous modes: $E_k(t, r) = \sqrt{2\pi\hbar\omega_k/\Omega\hat{\alpha}_k} e^{-i\omega_k t + ikr}$ and using the continuum limit: $\sum_k \rightarrow \int_{-\infty}^{\infty} \tilde{D}(\omega_k) \frac{d\omega_k}{2\pi}$. In this case the density of radiation modes is a slowly varying function of frequency: $\tilde{D}(\omega) = \Omega\omega^2/\pi^2 c^3$ which allows to take it outside of the integration with the appropriate replacement of the frequency by a resonant matter frequency. The signal (S16) - (S17) then reads

$$\begin{aligned}
R_B^{(i)}(t'_s, \omega'_s; t'_r, \omega'_r) &= \mathcal{D}^2(\omega_a) \mathcal{D}^2(\omega_b) \int_{-\infty}^{\infty} d\tau_p d\tau'_p d\tau_s d\tau_r e^{-i\omega'_s \tau_s - i\omega'_r \tau_r} \int_{-\infty}^{\infty} dt_p dt'_p \\
&\times \mathcal{E}_p^*(t_p - \tau_p - \bar{t}_p, r_a) \mathcal{E}_p(t_p - \bar{t}_p, r_a) \mathcal{E}_p^*(t'_p - \tau'_p, r_b) \mathcal{E}_p(t'_p, r_b) \\
&\times \langle g | \hat{V}(t_p - \tau_p) \hat{V}^\dagger(t'_r + \tau_r) \hat{V}(t'_r) \hat{V}^\dagger(t_p) | g \rangle_a \\
&\times \langle g | \hat{V}(t'_p - \tau'_p) \hat{V}^\dagger(t'_s + \tau_s) \hat{V}(t'_s) \hat{V}^\dagger(t'_p) | g \rangle_b,
\end{aligned} \tag{S18}$$

$$\begin{aligned}
R_B^{(ii)}(t'_s, \omega'_s; t'_r, \omega'_r) &= -\mathcal{D}^2(\omega_a) \mathcal{D}^2(\omega_b) \int_{-\infty}^{\infty} d\tau_p d\tau'_p d\tau_s d\tau_r e^{-i\omega'_s \tau_s - i\omega'_r \tau_r} \int_{-\infty}^{\infty} dt_p dt'_p \\
&\times \mathcal{E}_p^*(t_p - \tau_p - \bar{t}_p, r_a) \mathcal{E}_p(t_p - \bar{t}_p, r_a) \mathcal{E}_p^*(t'_p - \tau'_p, r_b) \mathcal{E}_p(t'_p, r_b) \\
&\times \langle g | \hat{V}(t_p - \tau_p) \hat{V}^\dagger(t'_r + \tau_r) \hat{V}(t'_s) \hat{V}^\dagger(t_p) | g \rangle_a \\
&\times \langle g | \hat{V}(t'_p - \tau'_p) \hat{V}^\dagger(t'_s + \tau_s) \hat{V}(t'_r) \hat{V}^\dagger(t'_p) | g \rangle_b,
\end{aligned} \tag{S19}$$

where $D(\omega) = 2\pi\tilde{D}(\omega)/\hbar\Omega$ and we changed the time variables of the pump pulse. We further assumed that the pulse is much longer than the dephasing time and extended the time integrations over τ_p, τ'_p, t_p and t'_p to infinity.

We next turn to the matter correlation functions in Eqs. (S18) - (S19). In the hole burning limit dephasing is short compare to fluctuation time scale and pump delay and detector central times $\tau_j \ll \bar{t}_j$, Λ_α^{-1} , $j = p, s, r$, $\alpha = a, b$. We further assume a Gaussian excitation pulse with bandwidth σ_p and central frequency ω_p : $\mathcal{E}_p(t) = \mathcal{E}_p e^{-\frac{1}{2}\sigma_p^2 t^2 - i\omega_p t}$. Assuming that the time and frequency gate bandwidths are broader than the inverse fluctuation time scale $\sigma_p, \sigma_T^j, \sigma_\omega^j \gg \Lambda_\alpha$, $j = r, s$, $\alpha = a, b$ one may neglect the fluctuations during the detection window and pulse duration such that $g_\alpha(t'_j) \simeq g_\alpha(\bar{t}_j)$, $j = r, s$, $g_\alpha(t_p) \simeq g_\alpha(0) = 0$. Expanding the linewidth functions to second order in τ_s, τ_r and τ_p and assuming that detector r clicks first: $t_r < t_s$ we evaluate the time integrals in Eqs. (S18) - (S19) using Eq. (S5) and obtain

$$R_B^{(i)}(t'_s, \omega'_s; t'_r, \omega'_r) = F_a^{(i)}(\bar{t}_r, \omega'_r) F_b^{(i)}(\bar{t}_s, \omega'_s), \tag{S20}$$

$$R_B^{(ii)}(t'_s, \omega'_s; t'_r, \omega'_r) = F_a^{(i)}(\bar{t}_r, \bar{t}_s, \omega'_r) F_b^{(ii)}(\bar{t}_r, \bar{t}_s, \omega'_s) e^{-i\omega_{ab}(t'_s - t'_r) - \tilde{g}_a(\bar{t}_r, \bar{t}_s) - \tilde{g}_b^*(\bar{t}_r, \bar{t}_s)}, \tag{S21}$$

where

$$F_\alpha^{(i)}(t, \omega) = \frac{\mathcal{D}^2(\omega_\alpha) |\mu_\alpha|^4 |\mathcal{E}_p|^2}{\sigma_p \tilde{\sigma}_{p\alpha} \tilde{\sigma}_{\alpha 0}(t)} \exp \left[-\frac{(\omega_p - \omega_\alpha)^2}{2\tilde{\sigma}_{p\alpha}^2} - \frac{(\omega - \tilde{\omega}_\alpha(t))^2}{2\tilde{\sigma}_{\alpha 0}(t)^2} \right], \tag{S22}$$

$$F_\alpha^{(ii)}(t_1, t_2, \omega) = \frac{\mathcal{D}^2(\omega_\alpha) |\mu_\alpha|^4 |\mathcal{E}_p|^2}{\sigma_p \sigma_{p\alpha}(t_1, t_2) \sigma_{\alpha 0}(t_1, t_2)} \exp \left[-\frac{(\omega_p - \omega_{p\alpha}(t_1, t_2))^2}{2\sigma_{p\alpha}^2} - \frac{(\omega - \omega_\alpha(t_1, t_2))^2}{2\sigma_{\alpha 0}(t_1, t_2)^2} \right], \alpha = a, b. \tag{S23}$$

Here $\tilde{g}_\alpha(t_1, t_2) = \frac{2i\lambda_\alpha}{\Lambda_\alpha} [F_\alpha(t_1) - F_\alpha(t_2)] + \left[\frac{\Delta_\alpha^2}{\Lambda_\alpha^2} + i\frac{\lambda_\alpha}{\Lambda_\alpha} \right] F_\alpha(t_2 - t_1)$ with $t_1 < t_2$ and the remaining parameters are listed in Section S4. Combining the bare spectrograms (S20) - (S21) with the detector spectrogram (S6) and using Eq. (9) we finally obtain Eq. (1).

S4. GATING AND SPECTRAL DIFFUSION PARAMETERS

In Eq. (1) - (3) the normalization functions for $\alpha = a, b$ and $j = r, s$:

$$C_{\alpha 0}^j(t) = \frac{\mathcal{D}^2(\omega_\alpha)|\mu_\alpha|^4|\mathcal{E}_p|^2}{\tilde{\sigma}_{p\alpha}\tilde{\sigma}_{\alpha 0}(t)[\tilde{\sigma}_{\alpha 0}^{-2}(t) + \sigma_{Dj}^{-2}]^{1/2}}, \quad I_{\alpha 0}^j(t_1, t_2) = \frac{\mathcal{D}^2(\omega_\alpha)|\mu_\alpha|^4|\mathcal{E}_p|^2}{\sigma_{p\alpha}^j(t_1, t_2)\sigma_{\alpha 0}^j(t_1, t_2)[(\sigma_{\alpha 0}^j)^{-2}(t_1, t_2) + \sigma_{Dj}^{-2}]^{1/2}}, \quad (\text{S24})$$

where $\mu_\alpha \equiv \mu_{\alpha g}$ and $\omega_\alpha \equiv \omega_\alpha - \omega_g = \omega_\alpha^0 + \lambda_\alpha$. Time dependent frequency shifts:

$$\tilde{\omega}_\alpha(t) = \omega_\alpha^0 - \lambda_\alpha + M_\alpha(t) \left[2\lambda_\alpha + \frac{\Delta_\alpha^2(\omega_p - \omega_a^0 - \lambda_a)}{\tilde{\sigma}_{p\alpha}^2} \right], \quad (\text{S25})$$

$$\omega_{p\alpha}^r(t_1, t_2) = \omega_\alpha^0 + \lambda_\alpha[1 - M_\alpha(t_2) + M_\alpha(t_1)], \quad \omega_{p\alpha}^s(t_1, t_2) = \omega_\alpha^0 + \lambda_\alpha[1 + M_\alpha(t_2) - M_\alpha(t_1)], \quad (\text{S26})$$

$$\begin{aligned} \omega_\alpha^r(t_1, t_2) &= \frac{1}{2}(\omega_a + \omega_b) - \lambda_\alpha M_\alpha(t_2 - t_1) + M_\alpha(t_1) \left[2\lambda_\alpha + \frac{\Delta_\alpha^2[\omega_p - \omega_{p\alpha}^r(t_1, t_2)]}{\sigma_{p\alpha}^2(t_1, t_2)} \right], \\ \omega_\alpha^s(t_1, t_2) &= \frac{1}{2}(\omega_a + \omega_b) + \lambda_\alpha M_\alpha(t_2 - t_1) + M_\alpha(t_2) \left[2\lambda_\alpha + \frac{\Delta_\alpha^2[\omega_p - \omega_{p\alpha}^s(t_1, t_2)]}{\sigma_{p\alpha}^2(t_1, t_2)} \right], \end{aligned} \quad (\text{S27})$$

$$\omega_{\tau\alpha}^j(t_1, t_2, \omega) = \omega + \frac{\sigma_\omega^{j2}}{\sigma_\alpha^{j2}(t_1, t_2)} [\omega_\alpha^j(t_1, t_2) - \omega], \quad (\text{S28})$$

where $M_\alpha(t) = e^{-\Lambda_\alpha t}$. Time dependent dispersions:

$$\tilde{\sigma}_{p\alpha}^2 = \frac{1}{2}\sigma_p^2 + \Delta_\alpha^2, \quad \tilde{\sigma}_\alpha^{j2}(t) = \tilde{\sigma}_{\alpha 0}^2(t) + \sigma_{Dj}^2, \quad \tilde{\sigma}_{\alpha 0}^2(t) = \Delta_\alpha^2 \left[1 - \frac{\Delta_\alpha^2 M_\alpha^2(t)}{\tilde{\sigma}_{p\alpha}^2} \right], \quad \sigma_{Dj}^2 = \frac{1}{2}\sigma_T^2 + \sigma_\omega^{j2}, \quad (\text{S29})$$

$$\begin{aligned} \sigma_{p\alpha}^2(t_1, t_2) &= \frac{1}{2}\sigma_p^2 + \Delta_\alpha^2[1 + M_\alpha(t_1) - M_\alpha(t_2)], \quad \sigma_{p\alpha}^{s2}(t_1, t_2) = \frac{1}{2}\sigma_p^2 + \Delta_\alpha^2[1 + M_\alpha(t_2) - M_\alpha(t_1)], \\ \sigma_\alpha^{j2}(t_1, t_2) &= \sigma_{Dj}^2 + \sigma_{\alpha 0}^{j2}(t_1, t_2), \\ \sigma_{\alpha 0}^2(t_1, t_2) &= \Delta_\alpha^2 \left[M_\alpha(t_2 - t_1) - \frac{\Delta_\alpha^2 M_\alpha(t_1)}{\sigma_{p\alpha}^2(t_1, t_2)} \right], \quad \sigma_{\alpha 0}^{s2}(t_1, t_2) = \Delta_\alpha^2 \left[M_\alpha(t_2 - t_1) - \frac{\Delta_\alpha^2 M_\alpha(t_2)}{\sigma_{p\alpha}^2(t_1, t_2)} \right], \end{aligned} \quad (\text{S30})$$

$$\sigma_{\tau\alpha}^{j2}(t_1, t_2) = \sigma_\omega^{j2} \left[1 - \frac{\sigma_\omega^{j2}}{\sigma_\alpha^{j2}(t_1, t_2)} \right]. \quad (\text{S31})$$

In the absence of fluctuations $\Lambda_\alpha = 0$ $\alpha = a, b$ for the identical detector $\sigma_\omega^r = \sigma_\omega^s = \sigma_\omega$ and $\sigma_T^r = \sigma_T^s = \sigma_T$ such that $\sigma_D^2 = \frac{1}{2}\sigma_T^2 + \sigma_\omega^2$ the coincidence counting signal (1) becomes (4) where

$$\Omega_\tau = \bar{\omega}_r - \bar{\omega}_s + \frac{\sigma_\omega^2}{\sigma_a^2}[\bar{\omega}_a - \bar{\omega}_r] - \frac{\sigma_\omega^2}{\sigma_b^2}[\bar{\omega}_b - \bar{\omega}_s], \quad \sigma_\tau^2 = \sigma_\omega^2 \left[2 - \frac{\sigma_\omega^2}{\sigma_a^2} - \frac{\sigma_\omega^2}{\sigma_b^2} \right], \quad (\text{S32})$$

$$\eta = \frac{\tilde{I}_a^r \tilde{I}_b^s + \tilde{I}_a^s \tilde{I}_b^r}{\tilde{C}_a^r \tilde{C}_b^s + \tilde{C}_a^s \tilde{C}_b^r}, \quad \tilde{C}_\alpha^j = e^{-\frac{(\bar{\omega}_j - \bar{\omega}_\alpha)^2}{2\sigma_a^2}}, \quad \tilde{I}_\alpha^j = e^{-\frac{(\bar{\omega}_j - \bar{\omega}_\alpha)^2}{2\sigma_a^2} - \frac{\omega_{ab}^2}{4\sigma_T^2}}, \quad j = r, s. \quad (\text{S33})$$

Here

$$\bar{\omega}_\alpha = \tilde{\omega}_\alpha + \frac{1}{2}(\omega_{\bar{\alpha}} - \omega_\alpha), \quad \tilde{\omega}_\alpha = \omega_\alpha + \frac{\Delta_\alpha^2(\omega_p - \omega_\alpha)}{\tilde{\sigma}_{p\alpha}^2}, \quad (\text{S34})$$

where $\bar{\alpha} = a$ if $\alpha = b$ and $\bar{\alpha} = b$ if $\alpha = a$,

$$\sigma_{\alpha}^2 = \sigma_D^2 + \Delta_{\alpha}^2 \left[1 - \frac{\Delta_{\alpha}^2}{\bar{\sigma}_{p\alpha}^2} \right]. \quad (\text{S35})$$

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- [1] Stolz, H. *Time-Resolved Light Scattering from Excitons*. (Springer-Verlag, Berlin, 1994).
 [2] Harbola, U. and Mukamel, S. Superoperator nonequilibrium Green's function theory of many-body systems; Applications to charge transfer and transport in open junctions *Phys. Rep.* **465**, 191 (2008).
 [3] Glauber, R. *Quantum Theory of Optical Coherence: Selected Papers and Lectures*. (Wiley, 2007).