Supporting Information

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SI Text

TF-Resolved Photon Counting for the Time-Dependent JC Model. In this section, we show the power of TF-resolved photon counting to the canonical JC model of quantum optics. We add one additional level of complexity by treating the cavity mode–atom as time dependent. The gating will then provide a versatile tool to study exciton dynamics through photon correlations. We will provide a simple analytical solution for a particular form of time-dependent cavity modulation.

The Propagator for the Time-Dependent JC Model. To describe the quantized radiation field, we introduce the Hermitian photon number operator A_0. Changes of the cavity photon number by m quanta are represented by creation and annihilation operators $A_\pm$, where $A_\pm^\dagger = A_\mp$, which satisfy the relation $A_0 A_\pm = A_\pm(A_0 \pm m)$, where m is a nonzero real number of quanta. They satisfy the commutation relations $[A_0, A_\pm] = \pm m A_\pm$. A_+A_-, which represents a process in which m quanta are first destroyed and then created, depends on the number of initial quanta A_0, such that $A_+A_- = \chi(A_0)$, where χ is the real function of its argument. Similarly, A_-A_+ should satisfy $A_-A_+ = \chi(A_0 + m)$. The operators A_0, A_+, and A_- form a closed oscillator algebra for any m-quanta ladder. We can construct a generalized time-dependent JC model Hamiltonian

$$H = r(A_0) + s(A_0)\sigma_z + \lambda(t)(A_+\sigma_- + A_-\sigma_+). \quad \textbf{[S1]}$$

For single-photon transition processes, such that $A_0 = a^\dagger a = n$, $r(n) = \omega n$, $s(n) = \omega_0/2$, and $\chi(\Delta) = \Delta$, Eq. **S1** reduces to Eq. **1**.

After the general algebra, one can split the Hamiltonian into $H = H_0 + H_i$, where $H_0 = \omega(\Delta)$ is the field energy function:

$$\omega(\Delta) = \frac{1}{2}[r(\Delta - m) + r(\Delta) + s(\Delta - m) - s(\Delta)], \quad \textbf{[S2]}$$

while $H_i = \delta(\Delta)\sigma_z + \lambda(t)(A_+\sigma_- + A_-\sigma_+)$, where the detuning function $\delta(\Delta)$ is given by

$$\delta(\Delta) = \frac{1}{2}[r(\Delta - m) - r(\Delta) + s(\Delta - m) + s(\Delta)]. \quad \textbf{[S3]}$$

We thus have the SU(2) algebra governed by $F_\pm$ and F_0, which connect with angular momentum algebra as

$$F_\pm = \pm J_\pm, \quad F_0 = 2J_z, \quad J_\pm = J_x \pm iJ_y. \quad \textbf{[S4]}$$

It is easy to see that $A_\pm\sigma_\mp$ satisfies

$$[A_-\sigma_+, A_+\sigma_-] = \chi(\Delta)\sigma_z, \quad \textbf{[S5]}$$

$$\{A_-\sigma_+, A_+\sigma_-\} = \chi(\Delta), \quad \textbf{[S6]}$$

which yields

$$F_0F_\pm = \pm F_\pm, \quad F_\pm F_0 = \mp F_\pm, \quad F_\pm F_\mp = -\frac{1}{2}(1 \pm F_0). \quad \textbf{[S7]}$$

We further have $F_0^2 = 1$, $F_\pm^2 = 0$. The time evolution operator can be recast as

$$U(t) = e^{-i\omega(\Delta)t}U_i(t), \quad \textbf{[S8]}$$

where $U_i(t)$ is the evolution operator governed by H_i. Using the SU(2) algebra, we can recast the Hamiltonian H_i as

$$H_i = \delta(\Delta)F_0 + \lambda(t)\sqrt{\chi(\Delta)}(F_+ - F_-). \quad \textbf{[S9]}$$

Following the Wei–Norman formalism [**S58**] and using the exponentiating rule of SU(2) algebra

$$e^{ia(\mathbf{n}\cdot\boldsymbol{\sigma})} = I\cos a + i(\mathbf{n}\cdot\boldsymbol{\sigma})\sin a \quad \textbf{[S10]}$$

for arbitrary a, using relations [**S7**], and neglecting $\mathcal{O}(f^2, g^2, fg)$, we obtain for the evolution operator

$$U_i(t) = \cosh[h(t)] + \sinh[h(t)]F_0 + \mathcal{G}(t)F_+ + \mathcal{F}(t)F_-, \quad \textbf{[S11]}$$

where $\mathcal{G}(t) = g(t)e^{h(t)}$, $\mathcal{F}(t) = f(t)e^{-h(t)}$. Unitarity implies that $\mathcal{G}(t) = \mathcal{F}^*(t)$. We also introduce the auxiliary function $\mathcal{H}(t) = e^{-h(t)}$. On resonant $\delta = 0$, functions $X = \mathcal{F}, \mathcal{H}$ satisfy Eq. **6** with initial conditions $\mathcal{H}(t_0) = 1$, $\dot{\mathcal{H}}(t_0) = 0$, $\mathcal{F}(t_0) = 0$, $\dot{\mathcal{F}}(t_0) = i\sqrt{n+1}\lambda(t_0) = -i\alpha/(2\tau)$. Changing the variable to

$$z(t) = \frac{e^{(t-T)/\tau}}{1 + e^{(t-T)/\tau}}, \quad \textbf{[S12]}$$

Eq. **6** reads

$$z(1-z)\frac{d^2X}{dz^2} + (\beta - z)\frac{dX}{dz} + \alpha^2 X = 0, \quad \textbf{[S13]}$$

where $\beta = 1/2$. This is the hypergeometric equation with $\beta = -\alpha$. The solution of Eq. **S13** for $\beta = 1/2$ as given by Eq. **7**.

Spontaneous Emission in an Imperfect Cavity. A two-level atom in the cavity can be described by a joint photon–atom wave function

$$|\psi\rangle = v(t)|\downarrow, 0\rangle + w(t)|\uparrow, 1\rangle, \quad \textbf{[S14]}$$

where the first argument in the bracket corresponds to the atomic state and the second represents the photon number. The total Hamiltonian $H = H_{JC} + H_{vac}$ contains the strong atom–cavity mode coupling governed by Eq. **1** and the coupling to vacuum noncavity modes

$$H_{vac} = V\sum_{\mathbf{k}} g_{\mathbf{k}} a_{\mathbf{k}}^\dagger + H.c.. \quad \textbf{[S15]}$$

The quantum master equation for the density operator including cavity modes is calculated to first order in H_{JC} and second order in H_{vac}:

$$\dot{\rho} = -\frac{i}{\hbar}[H_{JC}(t), \rho] - \frac{1}{\hbar^2}\int^t dt'[H_{vac}(t)[H_{vac}(t'), \rho(t')]] - \mathcal{L}_\kappa[\rho], \quad \textbf{[S16]}$$

where $\mathcal{L}_\kappa[\rho]$ represents the Lindblad operator responsible for cavity damping given by the rate κ. Using Markovian and Wigner–Weisskopf approximations, one can calculate the density operator assuming that $\rho_{\uparrow\uparrow}(0) = 1$, $\rho_{\downarrow\downarrow}(0) = 0$, and the ground-state population remains the same $\dot{\rho}_{\downarrow\downarrow} = 0$. We then obtain the excited-state population

$$\rho_{\uparrow\uparrow} \simeq e^{-2(\lambda_0^2/\kappa + \gamma_0)t}, \quad \textbf{[S17]}$$

where $\gamma_0 = \frac{\omega^3|\mu|^2}{3\hbar\epsilon_0\pi c^3}$ is the free space rate of spontaneous emission. Here, we assume that the solid angle extended by the cavity mirrors is very small and that the atomic decay rate into noncavity modes will then be comparable with the rate of atomic decay in free space. The bad cavity limit corresponds to the case when $\kappa \gg \lambda_0, \gamma_0$. The free space density of states is governed by $\rho_0 = \frac{\mathcal{V}\omega^2}{2\pi^2 c^3}$, while in the cavity,

$$\rho_{cav} = \frac{1}{\pi}\frac{\frac{\omega_c}{2Q}}{(\omega_c - \omega_k)^2 + \left(\frac{\omega_c}{2Q}\right)^2}. \quad \textbf{[S18]}$$

At atom–cavity resonance $\omega_k = \omega_c = \omega_0$, we obtain for the cavity decay in the bad cavity limit $2\lambda_0^2/\kappa \gg \gamma_{free}$:

$$\gamma_{cav} = 2\frac{\lambda_0^2}{\kappa\gamma_{free}} = \frac{2Q|\mu|^2}{\hbar\epsilon_0\mathcal{V}}. \quad \textbf{[S19]}$$

The Purcel factor is then defined as

$$f \equiv \frac{\gamma_{cav}}{\gamma_{free}} = \frac{3Q}{\mathcal{V}}\frac{2\pi c^3}{\omega_0^3}. \quad \textbf{[S20]}$$

From this expression, we can see that $f \gg 1$, and the photons are predominantly emitted into the cavity mode. One can, therefore, use perturbation theory with respect to the coupling to the noncavity mode and define the photon counting signals of the noncavity modes.

TF-Resolved Photon Counting. TF gated Nth-order photon correlation measurement performed at N detectors centered at time t_j and frequency ω_j, $j = 1, ..., N$ is defined as

$$g_{TF}^{(N)}(t_1,\omega_1,\Gamma_1; ..., t_N;\omega_N,\Gamma_N) = \langle \mathcal{T}\hat{n}_{t_1,\omega_1}...\hat{n}_{t_N,\omega_N}\rangle, \quad \textbf{[S21]}$$

where $g^{(1)}$ corresponds to a gated photon number, $g^{(2)}$ is a gated photon coincidence counting (PCC) signal, $\langle ...\rangle = \mathrm{Tr}[...\rho(t)]$, and $\rho(t)$ represents the full matter plus field density matrix and contains information about the system evolution before the detection (e.g., photon generation process, etc.). Γ_j, $j = 1, ...N$ represent other parameters of the detectors, such as bandwidth (Γ_{Tj} and $\Gamma_{\omega j}$ are the time gate and frequency gate bandwidths, respectively). The TF gated photon number superoperator is given by

$$\hat{n}_{t,\omega} = \int dt' \int d\tau D(t,\omega;t',\tau)\hat{n}(t',\tau). \quad \textbf{[S22]}$$

Here, $D(t,\omega,t',\tau)$ is a detector time domain spectrogram (ordinary function, not an operator), which takes into account the detector parameters

$$D(t,\omega,t',\tau) = \int \frac{d\omega''}{2\pi} e^{-i\omega''\tau}|F_f(\omega'',\omega)|^2 F_t^*(t'+\tau,t) \times F_t(t',t), \quad \textbf{[S23]}$$

where F_t and F_f are TF gating functions that are characterized by central time t, frequency ω, and detection bandwidths Γ_T and Γ_ω, respectively; $\hat{n}(t,t')$ is a bare photon number superoperator defined in terms of the bare field operators $a(t)$ as

$$\hat{n}(t',\tau) = \sum_{s,s'} \hat{E}_{sR}^\dagger(t'+\tau)\hat{E}_{s'L}(t')\rho(t'). \quad \textbf{[S24]}$$

Eq. **S22** can be alternatively recast in terms of Wigner spectrograms

$$\hat{n}_{t,\omega} = \int dt' \int \frac{d\omega'}{2\pi} W_D(t,\omega;t',\omega')\hat{n}(t',\omega'), \quad \textbf{[S25]}$$

where $W_D(t,\omega,t',\omega')$ is a detector Wigner spectrogram given by

$$W_D(t,\omega,t',\omega') = \int d\tau D(t,\omega,t',\tau)e^{i\omega'\tau} \quad \textbf{[S26]}$$

and the Wigner spectrogram for the bare photon number operator is given by

$$\hat{n}(t',\omega') = \int d\tau e^{-i\omega'\tau}\hat{n}(t',\tau). \quad \textbf{[S27]}$$

For Gaussian gates,

$$F_t(t',t) = e^{-\frac{1}{2}\Gamma_T^2(t'-t)^2}, \quad F_f(\omega',\omega) = e^{-\frac{(\omega'-\omega)^2}{4\Gamma_\omega^2}}, \quad \textbf{[S28]}$$

and the detector time domain and Wigner spectrograms are given by

$$D(t,\omega,t',\tau) = \frac{\Gamma_\omega}{\sqrt{2\pi}} e^{-\frac{1}{2}\Gamma_T^2(t'-t)^2 - \frac{1}{2}\tilde{\Gamma}_\omega^2\tau^2 - [\Gamma_T^2(t'-t)+i\omega]\tau} \quad \textbf{[S29]}$$

$$W_D(t,\omega;t',\omega') = N_D e^{-\frac{1}{2}\tilde{\Gamma}_T^2(t'-t)^2 - \frac{(\omega'-\omega)^2}{2\tilde{\Gamma}_\omega^2} - iA(\omega'-\omega)(t'-t)}, \quad \textbf{[S30]}$$

where

$$\tilde{\Gamma}_\omega^2 = \Gamma_T^2 + \Gamma_\omega^2, \quad \tilde{\Gamma}_T^2 = \Gamma_T^2 + \frac{1}{\Gamma_\omega^{-2}+\Gamma_T^{-2}},$$
$$N_D = \frac{1}{\Gamma^T[\Gamma_\omega^2+\Gamma_T^2]^{1/2}}, \quad A = \frac{\Gamma_T^2}{\Gamma_T^2+\Gamma_\omega^2}. \quad \textbf{[S31]}$$

Note that, although Γ_T and Γ_ω can be controlled independently, the actual TF resolution is controlled by $\tilde{\Gamma}_T$ and $\tilde{\Gamma}_\omega$, respectively, which satisfy Fourier uncertainty $\tilde{\Gamma}_\omega/\tilde{\Gamma}_T > 1$. Assuming Lorentzian gates

$$F_t(t',t) = \theta(t-t')e^{-\Gamma_T(t-t')}, \quad F_f(\omega',\omega) = \frac{i}{\omega'-\omega+i\Gamma_\omega}, \quad \textbf{[S32]}$$

the detector time domain and Wigner spectrograms are given by

$$D(t,\omega,t',\tau) = \frac{i}{2\Gamma_\omega}\theta(\tau)\theta(t'-t)e^{-(i\omega+\Gamma_\omega+\Gamma_T)\tau - 2\Gamma_T(t'-t)} \quad \textbf{[S33]}$$

$$W_D(t,\omega;t',\omega') = -\frac{1}{2\Gamma_\omega}\theta(t-t')\frac{e^{-2\Gamma_T(t'-t)}}{\omega'-\omega+i(\Gamma_T+\Gamma_\omega)}. \quad \textbf{[S34]}$$

In our calculations, we will use the latter, as it yields simpler and more transparent results.

Expressions for the gated spectrogram were derived earlier [**S39**]. The gated photon number operator is given by

$$\hat{n}_{t,\omega} = \int dt'' \hat{E}_{t,\omega}^\dagger(t'')\hat{E}_{t,\omega}(t''), \quad \textbf{[S35]}$$

where the gated field operator $\hat{E}_{t,\omega}(t'')$ is connected to the bare field operator $\hat{E}(t)$ as follows:

$$\hat{E}_{t,\omega}(t'') = \int_{-\infty}^{\infty} dt' F_f(t''-t',\omega)F_t(t',t)\hat{E}(t'). \quad \textbf{[S36]}$$

Here, we have assumed that the time gate is applied before the frequency gate. Similarly, if the frequency gate is applied first followed by the time gate, the gated field operator is given by

$$\hat{E}_{\omega,t}(t'') = \int_{-\infty}^{\infty} dt' F_t(t'',t)F_f(t''-t',\omega)\hat{E}(t'). \quad \textbf{[S37]}$$

As one can see from Eq. **S36**, the PS gating [**S40**] can be recovered by removing the time gate $F_t = 1$ and keeping an exponential frequency gate, such that

$$F_f(\omega,\omega') = \frac{i}{\omega'+\omega+i\Gamma_{ps}/2}. \quad \textbf{[S38]}$$

In this case, the PS photon coincidence signal is given by

$$g_{PS}^{(2)}(\omega_1,\omega_2;\tau) = \lim_{t\to\infty}\langle \hat{A}_{\omega_1,\Gamma_{ps1}}^\dagger(t)\hat{A}_{\omega_2,\Gamma_{ps2}}^\dagger(t+\tau) \times A_{\omega_2,\Gamma_{ps2}}(t+\tau)\hat{A}_{\omega_1,\Gamma_{ps1}}(t)\rangle, \quad \textbf{[S39]}$$

where

$$\hat{A}_{\omega,\Gamma_{ps}}(t) = \int_{-\infty}^{t} dt_1 e^{(i\omega-\Gamma_{ps}/2)(t-t_1)}\hat{E}(t_1) \quad \textbf{[S40]}$$

is a PS gated field. This form of the gated signal works for a stationary process (dependence on τ in the left-hand side rather than on t and τ, such that there are no integrals over t and τ). It also works if $t \gg \Gamma_{ps}^{-1}$, which means that Γ_{ps} cannot approach zero (perfect reflection in a Fabri Perot cavity). It also works when $\Gamma_{ps}\tau_0 \ll 1$, where τ_0 is the scale of change in the field envelope. For comparison, the TF PCC [**S21**] for $N = 2$ reads

$$g^{(2)}_{TF}(t_1, \omega_1, \Gamma_{T1}, \Gamma_{\omega1}; t_2; \omega_2, \Gamma_{T2}, \Gamma_{\omega2}) = \langle \mathcal{T} \hat{n}_{t_1,\omega_1} \hat{n}_{t_2,\omega_2} \rangle, \quad \textbf{[S41]}$$

which depends on two times t_1, t_2 and two frequency arguments ω_1, ω_2. Clearly, expression [**S41**] with the gating spectrograms [**S22**] and the bare signal [**S24**] is very general. First, independent control of TF gates, which guarantees Fourier uncertainty for the TF resolution along with the fact that bare photon number operator depends on two time variables $\hat{n}(t, \tau)$, allows us to capture any dynamical process down to very short-scale dynamics especially important for the ultrafast spectroscopy applications. Second, the versatile gating [**S22**] provides a unique tool that can capture nonequilibrium and nonstationary states of matter, which can be controlled by gating bandwidths. In this case, a series of frequency correlation plots for ω_1, ω_2 (keeping central frequency of the spectral gates as variables) for different time delays $t_1 - t_2$ yields a fully capable 2D spectroscopy tool capable of measuring ultrafast dynamics. Third, the superoperator expressions require time ordering and therefore, can be generalized on other correlation functions of the field operators that are not normally ordered. Superoperator algebra provides an effective tool for bookkeeping of field–matter interactions. Fourth, as we show in the next section, PCC can be recast in terms of matter correlation function by expanding the total density matrix operator in perturbation series and tracing the vacuum modes. This way, the photon counting measurement can be related to matter response, which is the standard treatment in nonlinear spectroscopy.

Connecting Photon Counting to Matter Correlations. To connect PCC signal to matter response, one needs to expand the density operator in Eq. **S24** in perturbative series over field–matter interactions:

$$\rho(t) = \mathcal{T} e^{-\frac{i}{\hbar}\int_{-\infty}^{t} d\tau' H'_{-}(\tau')}, \quad \textbf{[S42]}$$

where the Hamiltonian H'_{-} in the interaction picture in dipole and rotating wave approximation is given by

$$H'_{-}(t) = \hat{V}^\dagger(t)\hat{E}(t) + H.c., \quad \textbf{[S43]}$$

where $V^\dagger(V)$ represents raising (lowering) dipole operator, $\hat{E}(t) = \sum_s \sqrt{2\pi\hbar\omega_s/\mathcal{V}}\hat{a}_s e^{-i\omega_s t}$ is a field operator for vacuum modes, and $\mathcal{V}$ denotes mode quantization volume. We first calculate the TF-resolved photon number

$$n_{t,\omega} = \int dt' \int d\tau D(t, \omega; t', \tau) n(t', \tau), \quad \textbf{[S44]}$$

where bare photon number $n(t', \tau) \equiv \langle \mathcal{T}\hat{n}(t', \tau)\rangle$ is an expectation value of the bare photon number operator. The leading contribution is coming from second-order expansion of field–matter interactions with vacuum modes. The result yields

$$n(t', \tau) = \frac{1}{\hbar^2}\int_{-\infty}^{t'} dt_1 \int_{-\infty}^{t'+\tau} dt_2 \langle V^\dagger(t_2)\langle V(t_1)\rangle' \times \sum_{s,s'} \langle \hat{E}_{s'}(t_2)\hat{E}^\dagger_{s'}(t'+\tau)\hat{E}_s(t')\hat{E}^\dagger_s(t_1)\rangle, \quad \textbf{[S45]}$$

where we used superoperator time ordering, and $\langle ...\rangle' = \mathrm{Tr}[...\rho'(t)]$, where $\rho'(t)$ is the density operator that excludes vacuum modes. One can now evaluate explicitly the vacuum field correlation function, where $\hat{a}^\dagger_s(\hat{a}_s)$ is a creation (annihilation) operator for modes that satisfy boson commutation relation $[\hat{a}_s, \hat{a}_{s'}] = \delta_{s,s'}$. Replacing the discrete sum over modes by a continuous integral $\sum_s \to \frac{V}{(2\pi)^3}\int d\omega_s \tilde{D}(\omega_s)$ with $\tilde{D}(\omega_s)$ being the density of states, one can obtain

$$n(t', \tau) = \mathcal{D}^2(\omega)\langle V^\dagger(t'+\tau)V(t')\rangle', \quad \textbf{[S46]}$$

where $\mathcal{D}(\omega) = \frac{1}{2\pi}\tilde{D}(\omega)$ is a combined density of states evaluated at the central frequency of the detector ω for smooth-enough distribution of modes.

One can similarly calculate the second-order bare correlation function

$$\langle \mathcal{T}\hat{n}_{t_1,\omega_1}\hat{n}_{t_2,\omega_2}\rangle = \int dt'_1 \int d\tau_1 D^{(1)}(t_1\omega_1; t'_1, \tau_1) \times \int dt'_2 \int d\tau_2 D^{(2)}(t_2, \omega_2; t'_2, \tau_2) \times \langle \mathcal{T}\hat{n}(t'_1, \tau_1)\hat{n}(t'_2, \tau'_2)\rangle. \quad \textbf{[S47]}$$

The leading contribution to the bare PCC rate $\langle \mathcal{T}\hat{n}(t'_1, \tau_1)\hat{n}(t'_2, \tau'_2)\rangle$ is coming from fourth-order expansion over field–matter interactions

$$\langle \mathcal{T}\hat{n}(t'_1, \tau_1)\hat{n}(t'_2, \tau'_2)\rangle = \frac{1}{\hbar^4}\int_{-\infty}^{t'_1} dt_1 \int_{-\infty}^{t'_1+\tau_1} dt_3 \times \int_{-\infty}^{t'_2} dt_2 \int_{-\infty}^{t'_2+\tau_2} dt_4 \langle V^\dagger(t_4)V^\dagger(t_3)V(t_1)V(t_2)\rangle' \times \sum_{s,s'}\sum_{r,r'} \langle E_{r'}(t_4)E_{s'}(t_3)E^\dagger_{r'}(t'_2+\tau_2)E^\dagger_{s'}(t'_1+\tau_1) \times E_s(t'_1)E_r(t'_2)E^\dagger_s(t_1)E^\dagger_r(t_2)\rangle. \quad \textbf{[S48]}$$

After tracing back the vacuum modes, we obtain

$$\langle \mathcal{T}\hat{n}(t'_1, \tau_1)\hat{n}(t'_2, \tau'_2)\rangle = \mathcal{D}^2(\omega_1)\mathcal{D}^2(\omega_2) \times \langle V^\dagger(t'_2+\tau_2)V^\dagger(t'_1+\tau_1)V(t'_1)V(t'_2)\rangle'. \quad \textbf{[S49]}$$

Therefore, the fundamental material quantity that yields the emission spectra [**S44**] is a two-point dipole correlation function in Eq. **S46**, and for the coincidence $g^{(2)}$ measurement [**S41**], it is the four-point dipole correlation function in Eq. **S49**.

Evaluation of Matter Correlation Functions. One can use the algebra [**S7**] to show that

$$F_0|n,\uparrow\rangle = |n,\uparrow\rangle, \quad F_0|n,\downarrow\rangle = -|n,\downarrow\rangle, \quad F_+|n,\uparrow\rangle = 0 \quad F_+|n,\downarrow\rangle = |n-m,\uparrow\rangle, \quad F_-|n,\downarrow\rangle = 0, \quad F_-|n,\uparrow\rangle = -|n+m,\downarrow\rangle. \quad \textbf{[S50]}$$

We first calculate atomic inversion $\langle \sigma_z(t)\rangle' = \langle \psi_0|U(t) F_0 U^\dagger(t)|\psi_0\rangle$, which yields

$$\langle \sigma_z(t)\rangle' = \sum_n (|w_n|^2 - |v_{n+m}|^2)(1 - 2|\mathcal{F}_{n+m}(t)|^2) - 4\mathrm{Re}\sum_n w_n v^*_{n+m}\mathcal{H}_{n+m}(t)\mathcal{F}_{n+m}(t). \quad \textbf{[S51]}$$

Similarly, we calculate two-point [**S46**] and four-point [**S49**] correlation functions, which read as Eqs. **8** and **10**, respectively.

Limiting case: Time-independent coupling. In the case of time-independent coupling $\lambda(t) = \lambda_0$, the solution of differential Eq. **6** reads

$$\mathcal{H}_{n+m}(t) = \cos(\Omega_{n+m}t), \quad \mathcal{F}_{n+m}(t) = i\sin(\Omega_{n+m}t), \quad \textbf{[S52]}$$

where $\Omega_{n+m} = 2\lambda_0\sqrt{\chi(n+m)}$ is a Rabi frequency. In this case, inversion satisfies the famous JC result

$$\langle\sigma_z(t)\rangle' = \sum_n(|w_n|^2 - |v_{n+m}|^2)\cos(2\Omega_{n+m}t) - i\sum_n(w_n v_{n+m}^* - w_n^* v_{n+m})\sin(2\Omega_{n+m}t). \quad \textbf{[S53]}$$

The corresponding expressions (Eqs. **8–10**) involves

$$G_{in}(t) = w_n\cos(\Omega_{n+m}t) + iv_{n+m}\sin(\Omega_{n+m}t),$$
$$G_{cn}(t_i, t_j) = i\sin(\Omega_{n+m}(t_i - t_j)),$$
$$G_{pn}(t_i, t_j) = \cos(\Omega_{n+m}(t_i - t_j)). \quad \textbf{[S54]}$$

Since the coupling is static, one does not need the time gating. In this case, one can use the frequency gating characterized by frequency bandwidth Γ according to Eq. **S40** as in the work of del Valle and coworkers (51–53). Assuming that, initially, atom is in the ground state and the field probability distribution is governed by $p(n)$, we obtain for emission spectra

$$\langle\hat{A}^\dagger_{\omega,\Gamma}(t)\hat{A}_{\omega,\Gamma}(t)\rangle = \int_0^\infty d\tau_1\int_0^\infty d\tau_2 e^{[i(\omega_0-\omega)-\Gamma]\tau_1} \times e^{[i(\omega-\omega_0)-\Gamma]\tau_2}\langle V^\dagger(t-\tau_1)V(t-\tau_2)\rangle' \quad \textbf{[S55]}$$

$$\langle\hat{A}^\dagger_{\omega_1,\Gamma_1}(t_1)\hat{A}^\dagger_{\omega_2,\Gamma_2}(t_2)A_{\omega_2,\Gamma_2}(t_2)\hat{A}_{\omega_1,\Gamma_1}(t_1)\rangle = \int_0^\infty d\tau_1 d\tau_2 d\tau_3 d\tau_4 e^{[i(\omega_0-\omega_1)-\Gamma_1]\tau_1}e^{[i(\omega_1-\omega_0)-\Gamma_1]\tau_2} \times e^{[i(\omega_0-\omega_2)-\Gamma_2]\tau_3}e^{[i(\omega_2-\omega_0)-\Gamma_2]\tau_4}\langle V^\dagger(t_1-\tau_1)V^\dagger(t_2-\tau_3) \times V(t_2-\tau_4)V(t_1-\tau_2)\rangle', \quad \textbf{[S56]}$$

where the matter correlation functions are given by

$$\langle V^\dagger(t-\tau_1)V(t-\tau_2)\rangle' = \mathcal{D}^2(\omega)|\mu|^2\sum_{n=0}^\infty p(n) \times\cos\Omega_{n+m}(t-\tau_1)\cos\Omega_{n+m}(\tau_2-\tau_1)\cos\Omega_{n+m}(t-\tau_2) \quad \textbf{[S57]}$$

and

$$\langle V^\dagger(t_1-\tau_1)V^\dagger(t_2-\tau_3)V(t_2-\tau_4)V(t_1-\tau_2)\rangle' = \mathcal{D}^2(\omega_1)\mathcal{D}^2(\omega_2)|\mu|^4\sum_{n=0}^\infty p(n)\cos\Omega_{n+m}(t-\tau_1) \times\sin\Omega_{n+m}(t_2-\tau_3-t_1+\tau_1)\cos\Omega_{n+m}(\tau_4-\tau_3) \times\sin\Omega_{n+m}(t_2-\tau_4-t_1+\tau_2)\cos\Omega_{n+m}(t_2-\tau_2). \quad \textbf{[S58]}$$

Different states of light show different correlations. For instance, for thermal photons, the probability is given by $p_{th}(n) = \bar{n}_{th}^n/(\bar{n}_{th}+1)^{n+1}$, where the average number of thermal photons is given by $\bar{n}_{th} = [\exp(\hbar\omega/k_BT)-1]^{-1}$. For coherent state, $p_c(n) = \exp(-\bar{n}_c)\bar{n}^n/n!$ is Poissonian.

Qualitative discussion of the joint time/frequency resolution. Below, we present a qualitative analysis of the resolution in both PS and TF techniques. An interesting conclusion can be made just by comparing the gating spectrograms with the gated signals. To that end, one can recast the TF-resolved gated signal in the form that resembles the PS the most. One can recast Eqs. **S44** and **S55** as follows:

$$n_{PS}(t,\omega) = \int_0^\infty dt_1'\int_0^\infty dt_1'' e^{[i(\omega-\omega_0)-\Gamma/2]t_1'}e^{[i(\omega_0-\omega)-\Gamma/2]t_1''} \times\langle V^\dagger(t-t_1'')V(t-t_1')\rangle' \quad \textbf{[S59]}$$

for the PS and

$$n_{TF}(t,\omega) = \int_0^\infty dt_1'\int_0^\infty dt_1''\theta(t_1''-t_1')e^{[i(\omega-\omega_0)-\Gamma_-/2]t_1'} \times e^{[i(\omega_0-\omega)-\Gamma_+/2]t_1''}\langle V^\dagger(t-t_1'')V(t-t_1')\rangle' \quad \textbf{[S60]}$$

for the TF-resolved photon number. Eqs. **S59** and **S60** can be generally recast as Eq. **4**. Similarly, we obtain for the coincidence counting signal

$$g^{(2)}_{PS}(t_1,\omega_1;t_2,\omega_2) = \int_0^\infty dt_1'\int_0^\infty dt_1'' e^{[i(\omega_1-\omega_0)-\Gamma_1/2]t_1'}e^{[i(\omega_0-\omega_1)-\Gamma_1/2]t_1''} \times\int_0^\infty dt_2'\int_0^\infty dt_2'' e^{[i(\omega_2-\omega_0)-\Gamma_2/2]t_2'}e^{[i(\omega_0-\omega_2)-\Gamma_2/2]t_2''} \times\langle V^\dagger(t-t_1'')V^\dagger(t-t_2'')V(t-t_2')V(t-t_1')\rangle' \quad \textbf{[S61]}$$

for the PS and

$$g^{(2)}_{TF}(t_1,\omega_1;t_2,\omega_2) = \int_0^\infty dt_1'\int_0^\infty dt_1''\theta(t_1''-t_1')e^{[i(\omega_1-\omega_0)-\Gamma_{1-}/2]t_1'} \times e^{[i(\omega_0-\omega_1)-\Gamma_{1+}/2]t_1''}\int_0^\infty dt_2'\int_0^\infty dt_2''\theta(t_2''-t_2') \times e^{[i(\omega_2-\omega_0)-\Gamma_{2-}/2]t_1'}e^{[i(\omega_0-\omega_2)-\Gamma_{2+}/2]t_2''}\langle V^\dagger(t-t_1'') \times V^\dagger(t-t_2'')V(t-t_2')V(t-t_1')\rangle' \quad \textbf{[S62]}$$

for the TF-resolved PCC signal. Eqs. **S61** and **S62** can be generally recast as Eq. **5**.

Consider the asymptotic expansion of Eq. **7**, such that, in the zeroth order, one can approximate it for $t_0 = 0$ as

$$\mathcal{H}_n(t) \simeq (\cos(\pi\alpha_n) + [1-\cos(\pi\alpha_n)]e^{-(t-T_{hn+})/2\tau}) \times\theta(t-T_{hn+}) + [1-2e^{(t-T_{hn-})/2\tau}](1-\theta(t-T_{hn-})) + (-1)^{n+1}\cos(\tilde{\Omega}_n(t-T))\theta(t-T_{hn-})(1-\theta(t-T_{hn+})),$$
$$\mathcal{F}_n(t) \simeq i[(-\sin(\pi\alpha_n) + [1+\sin(\pi\alpha_n)]e^{-(t-T_{fn+})/2\tau}) \times\theta(t-T_{fn+}) - e^{(t-T_{fn-})/2\tau}(1-\theta(t-T_{fn-})) + (-1)^n\sin(\tilde{\Omega}_n(t-T))\theta(t-T_{fn-})(1-\theta(t-T_{fn+}))], \quad \textbf{[S63]}$$

where $\tilde{\Omega}_n = \pi\alpha_n/8\tau$ is a Rabi frequency, and

$$T_{hn-} = T - 16\tau k/\alpha_n, \quad T_{hn+} = T + 8\tau(1+2k/\alpha_n), \quad \textbf{[S64]}$$

$$T_{fn-} = T - 8\tau k/\alpha_n, \quad T_{fn+} = T + 8\tau(-1+2k/\alpha_n), \quad \textbf{[S65]}$$

where $k = 1, \pm1, \pm2, \ldots$. Performing time integrals in Eqs. **4** and **5** analytically with the asymptotic form of Eq. **S63**, we obtain a sum of several terms of generic form presented in the text.