# Two-photon-coincidence fluorescence spectra of cavity multi-polaritons; novel signatures of multiexciton generation. Supplimentary materials

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Here we calculate the eigenvectors and eigenvalues of the system Hamiltonian Eq. (1), which in the bosonized representation assumes the form:

$$\begin{aligned} H_{\mathrm{sys}} &= H_0 + H_1 \\ H_0 &= \hbar\omega_c a^\dagger a + \hbar\omega_{1S} b^\dagger b + \hbar G\left(a^\dagger b + b^\dagger a\right) \\ H_1 &= \hbar g b^\dagger b^\dagger b b - \hbar \nu G\left(a^\dagger b^\dagger b b + b^\dagger b^\dagger a b\right) \end{aligned} \tag{1}$$

The first term in $H_1$ deals with the deviation of the excitons from ideal bosons, thus correcting for the excitonic density effects. The second term describes an additional nonlinear interaction via the cavity mode.

It is convenient to express the solution of the non-perturbed Hamiltonian $H_0$ using Schwinger's angular momentum representation of bosonic operators.[?] The angular momentum operators are

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defined as:

$$2\hat{J}_x = a^\dagger b + b^\dagger a, \qquad 2i\hat{J}_y = a^\dagger b - b^\dagger a \tag{2}$$

$$2\hat{J}_z = a^\dagger a - b^\dagger b, \qquad \hat{J}^2 = \frac{\hat{N}}{2}\left(\frac{\hat{N}}{2}+1\right) \tag{3}$$

where $\hat{N} = a^\dagger a + b^\dagger b$ is the total number of particles operator.

The Hamiltonian can now be written in terms of SO(3) rotations

$$H_0 = \hbar\Omega\hat{N} + \hbar A e^{-i\theta\hat{J}_y}\hat{J}_z e^{i\theta\hat{J}_y} \tag{4}$$

Here we have introduced some auxiliary quantities

$$\Omega = \frac{1}{2}\left(\omega_C + \omega_{1S}\right) \qquad \Delta = \omega_C - \omega_{1S} \tag{5}$$

$$A = \sqrt{(\Delta^2 + 4G^2)} \qquad \tan\theta = \frac{2G}{\Delta} \tag{6}$$

For a fixed number $N$ of polaritons, the eigenvectors and eigenvalues of the above Hamiltonian are dressed exciton states:

$$\left|\psi^{(0)}_{j,m}\right\rangle = \exp\left(-i\theta\hat{J}_y\right)|j,m\rangle \tag{7}$$

$$E^{(0)}_{j,m} = \hbar\left(N\Omega + mA\right) \tag{8}$$

$$a|j,m\rangle = \sqrt{j+m}\,|j-1/2, m-1/2\rangle \tag{9}$$

$$b|j,m\rangle = \sqrt{j-m}\,|j-1/2, m+1/2\rangle \tag{10}$$

Here the common eigenstates of $\hat{J}^2$ and $\hat{J}_z$ are the Fock states with $j+m$ photons in the cavity and $j-m$ excitons in the quantum dot, respectively:

$$|j,m\rangle = \frac{\left(a^\dagger\right)^{j+m}\left(b^\dagger\right)^{j-m}}{\sqrt{(j+m)!\,(j-m)!}}|0\rangle \tag{11}$$

with $j = N/2$, and $m = N/2, \ldots, -N/2$. Using Baker-Hausdorff identity we obtain the matrix elements of the cavity photon and exciton in the polaritonic basis:

$$
\begin{gathered}
a^{\star}_{j,m;j',m'} = \langle \psi^{(0)}_{jm} | a^{\dagger} | \psi^{(0)}_{j'm'} \rangle = \\
\delta_{j',j-\frac{1}{2}} \left[ \delta_{m',m-\frac{1}{2}} \sqrt{j+m} \cos\frac{\theta}{2} - \delta_{m',m+\frac{1}{2}} \sqrt{j-m} \sin\frac{\theta}{2} \right] \\
b^{\star}_{j,m;j',m'} = \langle \psi^{(0)}_{jm} | b^{\dagger} | \psi^{(0)}_{j'm'} \rangle = \\
\delta_{j',j-\frac{1}{2}} \left[ \delta_{m',m+\frac{1}{2}} \sqrt{j-m} \cos\frac{\theta}{2} + \delta_{m',m-\frac{1}{2}} \sqrt{j+m} \sin\frac{\theta}{2} \right] \\
a_{j,m;j',m'} = \langle \psi^{(0)}_{jm} | a | \psi^{(0)}_{j'm'} \rangle = a^{\star}_{j',m';j,m} \\
b_{j,m;j',m'} = \langle \psi^{(0)}_{jm} | b | \psi^{(0)}_{j'm'} \rangle = b^{\star}_{j',m';j,m}
\end{gathered} \tag{12}
$$

Using Eq. (12) we can calculate perturbation part of the system Hamiltonian in the polariton basis:

$$
\begin{aligned}
&\langle \psi^{(0)}_{jm} | H_1 | \psi^{(0)}_{j'm'} \rangle = \\
&= \sum_{m1=-j'+1/2}^{j'-1/2} \sum_{m2=-j'+1}^{j'-1} \sum_{m3=-j'+1/2}^{j'-1/2} [\hbar g b^{\star}_{j,m;j'-1/2,m3} b^{\star}_{j'-1/2,m3;j'-1,m2} b_{j'-1,m2;j'-1/2,m1} b_{j'-1/2,m1;j',m'} \\
&\quad -\hbar\nu G a^{\star}_{j,m;j'-1/2,m3} b^{\star}_{j'-1/2,m3;j'-1,m2} b_{j'-1,m2;j'-1/2,m1} b_{j'-1/2,m1;j',m'} \\
&\quad -\hbar\nu G b^{\star}_{j,m;j'-1/2,m3} b^{\star}_{j'-1/2,m3;j'-1,m2} a_{j'-1,m2;j'-1/2,m1} b_{j'-1/2,m1;j',m'}] = \\
&= \hbar \delta_{j'j} \Big\{ \delta_{m'm} \left[ \frac{g}{4}(5j^2 - 3j + m^2) - m(2j-1)(g\cos\theta - \nu\sin\theta) - (j^2 + j - 3m^2)\left(\frac{g}{4}\cos 2\theta + \frac{\nu}{2}\sin 2\theta\right) \right] \\
&\quad +\delta_{m',m+1} \sqrt{j-m}\sqrt{j+m'} \left[ (2j-1)\left(\frac{g}{2}\sin\theta - \frac{\nu}{2}\cos\theta\right) - (2m'-1)\left(\frac{g}{4}\sin 2\theta - \frac{\nu}{2}\cos 2\theta\right) \right] \\
&\quad +\delta_{m',m-1} \sqrt{j-m'}\sqrt{j+m} \left[ (2j-1)\left(\frac{g}{2}\sin\theta - \frac{\nu}{2}\cos\theta\right) - (2m-1)\left(\frac{g}{4}\sin 2\theta - \frac{\nu}{2}\cos 2\theta\right) \right] \\
&\quad +\delta_{m',m+2} \sqrt{j-m}\sqrt{j+m+1}\sqrt{j-m'+1}\sqrt{j+m'} \left(\frac{g}{4}\sin^2\theta - \frac{\nu}{4}\sin 2\theta\right) \\
&\quad +\delta_{m',m-2} \sqrt{j-m'}\sqrt{j+m'+1}\sqrt{j-m+1}\sqrt{j+m} \left(\frac{g}{4}\sin^2\theta - \frac{\nu}{4}\sin 2\theta\right) \Big\}
\end{aligned} \tag{13}
$$

The perturbed Hamiltonian Eq. (1) are given by:

$$
\begin{aligned}
E_{j,m} =& E_{j,m}^{(0)} + \langle j,m|e^{i\theta J_y} H_1 e^{-i\theta J_y}|j,m\rangle = \hbar\Omega N + \hbar A m + \frac{1}{4}\hbar g(5j^2 - 3j + m^2) - \\
& - \frac{1}{4}(j^2 + j - 3m^2)(\hbar g \cos 2\theta + \hbar\nu \sin 2\theta) - m(2j-1)(\hbar g \cos\theta - \hbar\nu \sin\theta) \qquad (14)
\end{aligned}
$$

The corresponding eigenvectors have the form:

$$
|\psi_{j,m}\rangle = |\psi_{j,m}^{(0)}\rangle + \sum_{m1 \neq m} \frac{\langle \psi_{j,m}^{(0)}|H_1|\psi_{j,m1}^{(0)}\rangle}{E_{j,m}^{(0)} - E_{j,m1}^{(0)}} |\psi_{j,m1}^{(0)}\rangle \qquad (15)
$$

Utilizing Eq. (Eq. (13)) and Eq. (Eq. (15)) we can calculate the transition selection rules to the first order in parameters $g$ and $\nu$, obtaining:

$$
\mu_{j,m;j,m'}^{\star} = \mu_{1S}\langle \psi_{j,m}|B^{\dagger}|\psi_{j',m'}\rangle = \mu_{1S}\langle \psi_{j,m}^{(0)}|B^{\dagger}|\psi_{j',m'}^{(0)}\rangle + \mu_{1S} \sum_{m1 \neq m'} \frac{\langle \psi_{j',m1}^{(0)}|H_1|\psi_{j',m'}^{(0)}\rangle}{E_{j',m'}^{(0)} - E_{j',m1}^{(0)}} b_{j,m;j',m1}^{\star} \qquad (16)
$$

Here the unperturbed transition matrix element has the form:

$$
\langle \psi_{j,m}^{(0)}|B^{\dagger}|\psi_{j',m'}^{(0)}\rangle = b_{j,m;j',m'}^{\star} - \hbar\nu \sum_{m2=-j'}^{j'} \sum_{m1=-j'+1/2}^{j'-1/2} b_{j,m;j',m2}^{\star} b_{j',m2;j'-1/2,m1}^{\star} b_{j'-1/2,m1;j',m'} = \qquad (17)
$$

$$
\begin{aligned}
= \delta_{j',j-\frac{1}{2}} \Big\{ & \delta_{m',m+\frac{1}{2}} \sqrt{j-m} \cos\frac{\theta}{2} \left[ 1 + \frac{\nu}{2}(1 - 3j - m) + \frac{\nu}{2}(1 + j + 3m)\cos\theta \right] \\
& + \delta_{m',m-\frac{1}{2}} \sqrt{j+m} \sin\frac{\theta}{2} \left[ 1 + \frac{\nu}{2}(1 - 3j + m) - \frac{\nu}{2}(1 + j - 3m)\cos\theta \right] \\
& - \delta_{m',m+\frac{3}{2}} \sqrt{(j-m)(j-m-1)(j+m+1)} \frac{\nu}{2} \sin\theta \cos\frac{\theta}{2} \\
& - \delta_{m',m-\frac{3}{2}} \sqrt{(j+m)(j+m-1)(j-m+1)} \frac{\nu}{2} \sin\theta \sin\frac{\theta}{2} \Big\} \qquad (18)
\end{aligned}
$$

Apart from the specifics of the dephasing mechanism, the polariton eigenvalues Eq. (14) and transition dipole moments (Eq. (16)) are the only necessary ingredients for calculating the fluorescence signals with Sum-Over-States formalism. Using the loop diagrams in **??**, the single and two-photon fluorescence can be written as:

$$S^{(1)}(\omega_1) = \mathcal{N}\frac{2\pi\hbar\omega_1}{\Omega_1}\sum_{j0,m0}|\langle\psi(t_0)|\psi_{j0,m0}\rangle|^2\times \tag{19}$$
$$\times\,\mathtt{Im}\left[\frac{i}{\hbar}\langle\psi_{j0,m0}|B^\dagger\hat{G}^\dagger(E_{j0,m0}/\hbar-\omega_1)B|\psi_{j0,m0}\rangle\right]$$
$$S^{(3)}(\omega_1,\omega_2) = \mathcal{N}\frac{2\pi\hbar\omega_1}{\Omega_1}\frac{2\pi\hbar\omega_2}{\Omega_2}\sum_{j0,m0}|\langle\psi(t_0)|\psi_{j0,m0}\rangle|^2\times$$
$$\times\,\mathtt{Im}\,[\left(\frac{i}{\hbar}\right)^2\langle\psi_{j0,m0}|B^\dagger\hat{G}^\dagger(E_{j0,m0}/\hbar-\omega_1)B^\dagger\hat{G}^\dagger(E_{j0,m0}/\hbar-\omega_1-\omega_2)B\hat{G}(E_{j0,m0}/\hbar-\omega_1)B|\psi_{j0,m0}\rangle+$$
$$+\langle\psi_{j0,m0}|B^\dagger\hat{G}^\dagger(E_{j0,m0}/\hbar-\omega_2)B^\dagger\hat{G}^\dagger(E_{j0,m0}/\hbar-\omega_1-\omega_2)B\hat{G}(E_{j0,m0}/\hbar-\omega_1)B|\psi_{j0,m0}\rangle+\omega_1\leftrightarrow\omega_2]$$

Here $|\psi(t_0)\rangle$ is the initial state of the system,and $G(\omega)$ ($G^\dagger(\omega)$) is the retarded (advanced) propagator given by:

$$\hat{G}(E_{j0,m0}/\hbar+\omega) = i\hbar\sum_{jm}\frac{|\psi_{j,m}\rangle\langle\psi_{j,m}|}{\hbar\omega+E_{j0,m0}-E_{j,m}+i\hbar\gamma} \tag{20}$$

Using the transition selection rules in Eq. (16), and introducing notation $P(j0,m0)=|\langle\psi(t_0)|\psi_{j0,m0}\rangle|^2$ we obtain the final expressions for the signals in Eq. (19), Eq. (3).