Low-temperature spectroscopic studies of single molecules in 3-D and on 2-D hosts

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APPENDIX 1

The coincidence of photons on two detectors can be related to a steady-state solution of the following optical Bloch equations, where we ignore bunching due to the triplet state:[1]

$$\dot{\rho_{11}} = \gamma_{21}\rho_{22} - \frac{i\Omega}{2}\rho_{12} + \frac{i\Omega}{2}\rho_{21} \tag{A.1}$$

$$\dot{\rho_{22}} = -\gamma_{21}\rho_{22} + \frac{i\Omega}{2}\rho_{12} - \frac{i\Omega}{2}\rho_{21} \tag{A.2}$$

$$\dot{\rho_{12}} = -\frac{i\Omega}{2}\rho_{11} + \frac{i\Omega}{2}\rho_{22} - \pi\Gamma_0\rho_{12} \tag{A.3}$$

$$\dot{\rho_{21}} = \frac{i\Omega}{2}\rho_{11} - \frac{i\Omega}{2}\rho_{22} - \pi\Gamma_0\rho_{21}. \tag{A.4}$$

The term Ω is the Rabi frequency, $\pi\Gamma_0$ is the decoherence rate and γ_{21} is the depopulation rate of the excited state. The series of coupled differential equations can be solved using the Laplace transform with the condition that the population initiates from the ground state. To relate the solution for the population of the excited state ρ_{22} to an experimental photon-coincidence histogram, we need to derive the steady-state solution of ρ_{22}:

$$g^{(2)}(\tau) = \rho_{22} / \lim_{\tau\to\infty} \rho_{22}. \tag{A.5}$$

Without approximations and solved with MATLAB, the theoretical solution for $g^{(2)}(\tau)$ is of the following form:

$$g^{(2)}(\tau) = 1 - e^{-\frac{|\tau|}{2}(\pi\Gamma_0+\gamma_{21})}[\cosh{(\tilde{\Omega}|\tau|)} + (\frac{\pi\Gamma_0+\gamma_{21}}{2\tilde{\Omega}})\sinh{(\tilde{\Omega}|\tau|)}], \tag{A.6}$$

where $\tilde{\Omega}$ is given by:

$$\tilde{\Omega} = \frac{1}{2}\sqrt{(\pi\Gamma_0)^2 - 2\pi\Gamma_0\gamma_{21} - 4\Omega^2 + \gamma_{21}^2}. \tag{A.7}$$

Rewriting $\tilde{\Omega} = \frac{1}{2} i \sqrt{4\Omega^2 - \gamma_{21}^2 - (\pi\Gamma_0)^2 + 2\pi\Gamma_0\gamma_{21}}$ and using the identities sinh $(\tilde{\Omega}) = -i$ sin $(i\tilde{\Omega})$ and cosh $(\tilde{\Omega}) =$ cos $(i\tilde{\Omega})$ we obtain the more common solution:[2]

$$g^{(2)}(\tau) = 1 - e^{-\frac{|\tau|}{2}(\pi\Gamma_0 + \gamma_{21})}[\cos(\tilde{\Omega}|\tau|) + (\frac{\pi\Gamma_0 + \gamma_{21}}{2\tilde{\Omega}})\sin(\tilde{\Omega}|\tau|)], \qquad \text{(A.8)}$$

where the new $\tilde{\Omega}$ is $\frac{1}{2}\sqrt{4\Omega^2 - \gamma_{21}^2 - (\pi\Gamma_0)^2 + 2\pi\Gamma_0\gamma_{21}}$. When the two-level system is strongly excited, the magnitude of the Rabi frequency exceeds all other terms inside the square root and the term $\tilde{\Omega}$ in the cosine and sine term reduces to the Rabi frequency Ω itself.

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