Note for *Quantum Optics*: Quantum theory of Fluorescence

Ray-Kuang Lee[1]
Institute of Photonics Technologies, National Tsing Hua University, Hsinchu, 300, Taiwan
(Dated: Fall, 2021)

Reference:
Ch. 8, 9, 10 in *"Quantum Optics,"* by M. Scully and M. Zubairy.
Ch. 7 in *"Mesoscopic Quantum Optics,"* by Y. Yamamoto and A. Imamoglu.
Ch. 8 in *"The Quantum Theory of Light,"* by R. Loudon.
Ch. 14, 15 in *"Elements of Quantum Optics,"* by P. Meystre and M. Sargent III.
Ch. 8 in *"Introductory Quantum Optics,"* by C. Gerry and P. Knight.

I. MOLLOW'S TRIPLET: RESONANCE FLUORESCENCE SPECTRUM

Consider a two-level system driving by a classical field, with the following Hamiltonian, i.e., Jaynes-Cummings model:

$$H = \frac{\hbar}{2}\omega_a\sigma_z + \hbar\sum_k \omega_k a_k^\dagger a_k + \frac{\Omega}{2}\hbar(\sigma_- e^{i\omega_L t} + \sigma_+ e^{-i\omega_L t}) \quad (1)$$

$$+ \hbar\sum_k (g_k\sigma_+ a_k + g_k^* a_k^\dagger \sigma_-). \quad (2)$$

Here, we want to solve the generalized Bloch equations:

$$\dot\sigma_-(t) = i\frac{\Omega}{2}\sigma_z(t)e^{-i\Delta t} + \int_{-\infty}^{t} dt' G(t-t')\sigma_z(t)\sigma_-(t') + n_-(t) \quad (3)$$

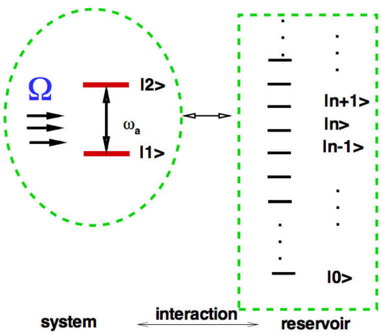
$$\dot\sigma_+(t) = -i\frac{\Omega}{2}\sigma_z(t)e^{i\Delta t} + \int_{-\infty}^{t} dt' G_c(t-t')\sigma_+(t')\sigma_z(t) + n_+(t) \quad (4)$$

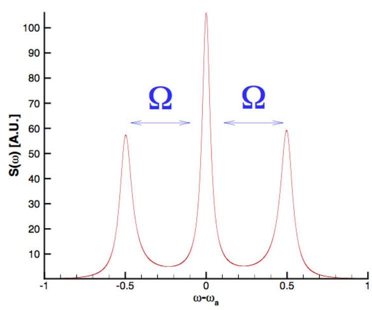
$$\dot\sigma_z(t) = i\Omega(\sigma_-(t)e^{i\Delta t} - \sigma_+(t)e^{-i\Delta t}) + n_z(t) \quad (5)$$

$$- 2\int_{-\infty}^{t} dt'[G(t-t')\sigma_+(t)\sigma_-(t') + G_c(t-t')\sigma_+(t')\sigma_-(t)] \quad (6)$$

Here, the coupling constant is defined as

$$g_k \equiv g_k(\hat{\mathbf{d}}, \overrightarrow{r}_0) = |d|\omega_a\sqrt{\frac{1}{2\hbar\epsilon_0\omega_k V}}\hat{\mathbf{d}}\cdot\mathbf{E}_k^*(\overrightarrow{r_0}), \quad (7)$$





with the memory functions:

$$G(\tau) \equiv \sum_k |g_k|^2 e^{i\Delta_k t}\Theta(\tau) \quad (8)$$

$$G_c(\tau) \equiv \sum_k |g_k|^2 e^{-i\Delta_k t}\Theta(\tau). \quad (9)$$

Within the Markovian approximation, we have

$$G(t) = G_c(t) = \Gamma\delta(t). \quad (10)$$

The corresponding *quantum noise operators* defined above are

$$n_-(t) = i\sum_k g_k e^{i\Delta_k t}\sigma_z(t)a_k(-\infty) \quad (11)$$

$$n_+(t) = -i\sum_k g_k^* e^{-i\Delta_k t}a_k^+(-\infty)\sigma_z(t) \quad (12)$$

$$n_z(t) = 2i\sum_k [g_k^* e^{-i\Delta_k t}a_k^+(-\infty)\sigma_-(t) - g_k e^{i\Delta_k t}\sigma_+(t)a_k^+(-\infty)] \quad (13)$$

where the mean and the correlation functions of the reservoir before interaction:

$$< a_k(-\infty) >_R = < a_k^\dagger(-\infty) >_R = 0 \quad (14)$$

$$< a_k(-\infty)a_{k'}(-\infty) >_R = 0 \quad (15)$$

$$< a_k^\dagger(-\infty)a_{k'}^\dagger(-\infty) >_R = 0 \quad (16)$$

$$< a_k^\dagger(-\infty)a_{k'}(-\infty) >_R = \bar{n}_k\delta_{kk'} \quad (17)$$

$$< a_k(-\infty)a_{k'}^\dagger(-\infty) >_R = (\bar{n}_k + 1)\delta_{kk'}. \quad (18)$$

A. Fluorescence spectrum

in the frequency domain, solutions for the optical Bloch equations are:

$$\tilde{\sigma}_-(\omega+\Delta) = \frac{(2g\,h+\Omega^2)\,\tilde{n}_-(\omega) + \Omega^2\,\tilde{n}_+(\omega) + i\Omega g\,\tilde{n}_z(\omega) - i2\pi\Omega g[\tilde{G}(\omega)+\tilde{G}_c(\omega)]\delta(\omega)}{\Omega^2(f+g)+2f\,g\,h} \quad (19)$$

$$\tilde{\sigma}_+(\omega-\Delta) = \frac{\Omega^2\,\tilde{n}_-(\omega) + (2f\,h+\Omega^2)\,\tilde{n}_+(\omega) - i\Omega f\,\tilde{n}_z(\omega) + i2\pi\Omega f[\tilde{G}(\omega)+\tilde{G}_c(\omega)]\delta(\omega)}{\Omega^2(f+g)+2f\,g\,h} \quad (20)$$

$$\tilde{\sigma}_z(\omega) = \frac{2i\Omega g\,\tilde{n}_-(\omega) - 2i\Omega f\,\tilde{n}_+(\omega) + 2f\,g\,\tilde{n}_z(\omega) - 4\pi f\,g[\tilde{G}(\omega)+\tilde{G}_c(\omega)]\delta(\omega)}{\Omega^2(f+g)+2f\,g\,h} \quad (21)$$

where

$$f(\omega) = -i\omega - i\Delta + \tilde{G}(\omega)$$
$$g(\omega) = -i\omega + i\Delta + \tilde{G}_c(\omega)$$
$$h(\omega) = -i\omega + \tilde{G}(\omega) + \tilde{G}_c(\omega).$$

For the two-time correlation function of the atomic dipole is proportional to the first order correlation function $g^{(1)}(\tau)$, we can obtain the fluorescence spectrum by taking the Fourier transform of the first order correlation function:

$$S(\omega) = \int_{-\infty}^{\infty} d\tau\, g^{(1)}(\tau)e^{i\omega\tau} \propto \langle\tilde{\sigma}_+(\omega)\tilde{\sigma}_-(-\omega)\rangle_R. \quad (22)$$

It should be noted that here we cannot directly apply the *quantum regression theorem* since it is invalid for non-Markovian process.

At free space, one can assume the memory functions are delta functions since $\sum_k |g_k|^2 e^{i\Delta_k t} = \Gamma\delta(t)$ with Γ being the decay rate of the excited atom. Then, the noise correlation functions at zero temperature are also delta-function correlated (i.e., white noises). Therefore, the fluorescence spectrum at steady state is given by:

$$\langle \tilde{\sigma_+}(\omega)\tilde{\sigma_-}(-\omega)\rangle_R = \frac{\pi^2\Omega^2(\frac{\Gamma^2}{4}+\Delta^2)}{\frac{\Omega^2}{2}+\Delta^2+\frac{\Gamma^2}{4}}\delta(\omega+\Delta) \tag{23}$$
$$+\frac{\pi\Gamma\Omega^4(\frac{\Omega^2}{2}+\Gamma^2+(\omega+\Delta)^2)}{2(\frac{\Omega^2}{2}+\Delta^2+\frac{\Gamma^2}{4})[\Gamma^2(\frac{\Omega^2}{2}+\Delta^2+\frac{\Gamma^2}{4}-2(\omega+\Delta)^2)^2+(\omega+\Delta)^2(\Omega^2+\Delta^2+\frac{5}{4}\Gamma^2-(\omega+\Delta)^2)^2]}$$

In the limit of strong on-resonance pumping ($\Omega \gg \Gamma$, $\Delta = 0$), Eq.(23) can be reduced to:

$$\langle \tilde{\sigma_+}(\omega)\tilde{\sigma_-}(-\omega)\rangle_R = 2\pi\{2\pi\frac{\Gamma^2}{4\Omega^2}\delta(\omega) + \frac{\frac{3}{16}\Gamma}{(\omega+\Omega)^2+\frac{9}{16}\Gamma^2} + \frac{\frac{1}{4}\Gamma}{\omega^2+\frac{1}{4}\Gamma^2} + \frac{\frac{3}{16}\Gamma}{(\omega-\Omega)^2+\frac{9}{16}\Gamma^2}\} \tag{24}$$

Then, the resonance fluorescence spectrum exhibits the Mollow triplets for white noise: three Lorentzian profiles with peaks in the ratio $1:3:1$, and widths of $\frac{3}{2}\Gamma$, Γ, and $\frac{3}{2}\Gamma$.