Many atom fluorescence spectrum

Notations and computations largely taken from the phd thesis of Arthur Goetschy. We try here to extend the computations to a degenerate two-level system.

1 Equations of motions in the atomic basis

For a dipolar transition of moment μ_d, let us define a vectorial operator to represent the adimensioned dipole of atom i as $\mathbf{D}_i = \mathbf{D}_i^+ + \mathbf{D}_i^-$ with

$$\mathbf{D}_i^+ = \sum_{m\in\{-1,0,1\}} |e_{im}\rangle\langle g_i|\hat{\mathbf{e}}_m = (\mathbf{D}_i^-)^\dagger$$

These atoms interacts with the vacuum modes of the field, decomposed in the basis of plane waves $\{\mathbf{k},\boldsymbol{\varepsilon}\}$. We consider an addition external coherent pump at a drive frequency ω_d and modeled by a classical field

$$\mathbf{E}_d^\pm(\mathbf{r}_i) = E_d\boldsymbol{\varepsilon}_d e^{\pm i(\mathbf{k}_d\cdot\mathbf{r}-\omega_d t)}$$

The Hamiltonian of the complete system is

$$H = \hbar\omega_0\sum_i^{N_{\rm at}}\sum_{m\in\{-1,0,1\}} |e_{im}\rangle\langle e_{im}| + \hbar\sum_{k,\varepsilon}(a_{k,\varepsilon}^\dagger a_{k,\varepsilon}+1/2) - \sum_i \mathbf{D}_i\cdot\mathbf{E}_\perp(\mathbf{r}_i) + H_c$$

where we introduced a contact Hamiltonian $H_c = \sum_{i\neq j}\mathbf{D}_i\cdot\mathbf{D}_j\delta(\mathbf{r}_i-\mathbf{r}_j)/\varepsilon_0$ used to regularize later the divergence of the Green's function that we introduce in $\mathbf{E}_\perp$

$$\mathbf{E}_\perp^+(\mathbf{r}_i) = \mathbf{E}_0^+(\mathbf{r}_i) + \frac{\hbar\Gamma_0}{2\mu_d}\sum_j \mathbf{G}_{ij}\cdot\mathbf{D}_j^-$$

with the decay rate into the vacuum modes of the field

$$\Gamma_0 = \frac{\mu_d^2 k_0^3}{3\pi\varepsilon_0\hbar},$$

the vacuum field $\mathbf{E}_0$ and the Green's function of the Helmholtz equation

$$(\nabla\times\nabla - k_0^2)\boldsymbol{\mathcal{G}}(\mathbf{r};\omega_0) = \mathbb{1}\delta(\mathbf{r})$$

$$\boldsymbol{\mathcal{G}}(\mathbf{r};\omega_0) = \frac{\delta(\mathbf{r})}{3k_0^2}\mathbb{1}_3 - \frac{e^{ik_0 r}}{4\pi r}\Big(P(ik_0 r)\mathbb{1}_3 + Q(ik_0 r)\frac{\mathbf{r}\otimes\mathbf{r}}{r^2}\Big) \equiv -\frac{k_0}{6\pi}\mathbf{G}(\mathbf{r};\omega_0)$$

We then rewrite the Hamiltonian within the rotating wave approximation and where we canceled the diverging part with the contact Hamiltonian (and wrote the free field Hamiltonian as H_f)

$$H = \hbar(\omega_0 - i\Gamma_0/2)\sum_{i,m}\mathbf{D}_i^+\mathbf{D}_i^- - \sum_{i,\pm}\mathbf{D}_i^\pm\cdot\mathbf{E}_0^\pm(\mathbf{r}_i) - \frac{\hbar\Gamma_0}{2}\sum_{i\neq j}(\mathbf{D}_i^+\cdot\mathbf{G}_{ij}\cdot\mathbf{D}_j^- + h.c.) + H_f$$

We then compute the equation of motion of the m-th component $D_{im}^+ = \mathbf{D}_i^+\cdot\hat{\mathbf{e}}_m$ using $[D_{im}^+, D_{jn}^-] \propto \delta_{ij}$ and $[\mathbf{D}_i^\pm, \mathbf{E}_\perp^\pm(\mathbf{r}_i)] = 0$.

$$\begin{aligned}\frac{d}{dt}D_{im}^+ &= \frac{i}{\hbar}[H, D_{im}^+] \\ &= -i(\omega_0 - i\Gamma_0/2)\sum_n D_{in}^+[D_{im}^+, D_{in}^-] + \frac{i\Gamma_0}{2}\sum_{j\neq i,n,p} G_{np}^*(r_{ij})D_{jp}^+[D_{im}^+, D_{in}^-] - \frac{i\Gamma_0}{2}\sum_n \Omega_{in}^-[D_{im}^+, \\ &\quad D_{in}^-] + \mathcal{F}_{im}^+\end{aligned}$$

where we defined

- the adimensioned driving field (« classical » part of the field)

$$\mathbf{\Omega}_i^{\pm} = -2\frac{\mu_d}{\hbar\Gamma_0}E_d\boldsymbol{\varepsilon}_d e^{i\mathbf{k}_d\cdot\mathbf{r}}$$

- the quantum Langevin forces representing the measurement noise as a stochastic term when expectation values are taken with respect to the vacuum modes (« quantum » part of the field)

$$\mathcal{F}_{im}^{+} = -\frac{i}{2}\Omega_{0,m}^{-}(\mathbf{r}_i,t)[D_{im}^{+},D_{im}^{-}](t)e^{-i\omega_p t}$$

We need to compute the commutators appearing in the equations

$$\begin{aligned} D_{in}^{+}[D_{im}^{+},D_{in}^{-}] &= 0-|e_{in}\rangle\langle g_i|g_i\rangle\langle e_{in}|e_{im}\rangle\langle g_i| \\ &= -D_{im}^{+}\delta_{nm} \end{aligned}$$

We note that,

$$\begin{aligned} \{D_{im}^{+},D_{im}^{-}\} &= \mathbb{1}-\sum_{n\neq m}D_{in}^{+}D_{in}^{-} \\ [D_{im}^{+},D_{im}^{-}] &= D_{im}^{+}D_{im}^{-}+\sum_{n}D_{in}^{+}D_{in}^{-}-\mathbb{1} \end{aligned}$$

Such that we can rewrite

$$[D_{im}^{+},D_{jn}^{-}] = \delta_{ij}\left(D_{im}^{+}D_{in}^{-}+\delta_{nm}\left(\sum_{p}D_{ip}^{+}D_{ip}^{-}-\mathbb{1}\right)\right)$$

Finally we introduce the regularized Green's function (where the diverging real part has been canceled out by H_c and only the imaginary part remains in $\mathbf{r}=\mathbf{0}$)

$$G_{mn}(\mathbf{r}_{\mathrm{ij}}) = (1-\delta_{ij})\hat{\mathbf{e}}_m\cdot\mathbf{G}(\mathbf{r}_i-\mathbf{r}_j)\cdot\hat{\mathbf{e}}_n+i\delta_{ij}\delta_{nm}$$

One can finally obtain in the frame of the drive and with time and detuning in units of Γ_0^{-1},

$$\begin{aligned} \frac{d}{dt}D_{im}^{+} &= -i\Delta D_{im}^{+}+\frac{i}{2}\sum_{j,n,p}\left[(G_{ij}^{mn})^{*}D_{jn}^{+}D_{ip}^{+}D_{ip}^{-}+(G_{ij}^{pn})^{*}D_{jn}^{+}D_{im}^{+}D_{ip}^{-}\right]-\frac{i}{2}\sum_{j,n}(G_{ij}^{mn})^{*}D_{jn}^{+}- \\ &\quad \frac{i}{2}\sum_{p}(\Omega_{im}^{-}D_{ip}^{+}D_{ip}^{-}+\Omega_{ip}^{-}D_{im}^{+}D_{ip}^{-})+\frac{i}{2}\Omega_{im}^{-}+\mathcal{F}_{im}^{+} \\ \frac{d}{dt}D_{im}^{-} &= i\Delta D_{im}^{-}-\frac{i}{2}\sum_{j,n,p}\left[G_{ij}^{mn}D_{ip}^{+}D_{ip}^{-}D_{jn}^{-}+G_{ij}^{pn}D_{ip}^{+}D_{im}^{-}D_{jn}^{-}\right]+\frac{i}{2}\sum_{j,n}G_{ij}^{mn}D_{jn}^{-}+ \\ &\quad \frac{i}{2}\sum_{p}(\Omega_{im}^{+}D_{ip}^{+}D_{ip}^{-}+\Omega_{ip}^{+}D_{ip}^{+}D_{im}^{-})-\frac{i}{2}\Omega_{im}^{+}+\mathcal{F}_{im}^{-} \end{aligned}$$

Note that it is the elimination of terms of the form $D_{im}^{-}D_{im}^{+}$ using the closure relation on the single atom subspace that introduces couplings between polarization of the form $\sum_p D_{ip}^{+}D_{ip}^{-}$, which represents the population of atom i.

Reformulation as spins with long range interactions

This problem is equivalent to a fully connected spin problem with long range interactions because the raising and lowering operators follow the same algebra as spin operators $(|e_{im}\rangle\langle g_i|)^{(\dagger)} = \sigma_{im}^{+(-)}$ with the peculiarity that spins operators associated to the same atom but different polarizations are not independant, requiring two indices: $\sigma_{im}^{+}\sigma_{jn}^{+} \propto (1-\delta_{ij})$ and $\sigma_{im}^{+}\sigma_{in}^{-} = |e_{im}\rangle\langle e_{in}|$. To show it explicitely, we leave the driving field out for now and trace out over the degrees of freedom of the vacuum field to obtain, using the same approximations as before, obtaining the following master equation

$$\frac{d}{dt}\rho = \frac{i}{\hbar}[H_{\text{eff}}\rho - \rho H_{\text{eff}}^{\dagger}] + \sum_{ijnm} \gamma_{ij}^{nm}\sigma_{im}^{-}\rho\sigma_{jn}^{+}$$

where we introduced a non-Hermitian effective Hamiltonian that encompasses all but one of the terms

$$H_{\text{eff}} = \sum_{ijnm} J_{ij}^{mn}\sigma_{im}^{+}\sigma_{jn}^{-},$$

$$J_{ij}^{mn} = \hbar\omega_0\delta_{ij}\delta_{nm} - \frac{\hbar\Gamma_0}{2}G_{nm}(\mathbf{r}_{ij}),$$

and

$$\gamma_{ij}^{nm} = \Gamma_0 \text{Im}[G_{nm}(\mathbf{r}_{ij})].$$

We can rewrite $J_{ij}^{mn} = \hbar(\omega_0 - i\Gamma_0/2)\delta_{ij}\delta_{nm} - (1-\delta_{ij})\frac{\hbar\Gamma_0}{2}G_{nm}(\mathbf{r}_{ij})$ to recall that for a single atom, this procedure simply amounts to take into account the finite life time of the excited states.

2 Fluorescence spectrum

Refer to [Goetschy] for derivation and notations

$$I(r_d,t) = \langle \mathbf{\Omega}^{-}(r_d,t)\cdot\mathbf{\Omega}^{+}(r_d,t)\rangle = \sum_{i,j} \text{Tr}_3[\mathbf{G}^{*}(\mathbf{r}_d - \mathbf{r}_i)\langle \mathbf{D}_i^{+}\otimes\mathbf{D}_j^{-}\rangle(t)\mathbf{G}(\mathbf{r}_d - \mathbf{r}_j)]$$

$$\mathbf{\Omega}^{+}(r_d,\omega) = \mathbf{\Omega}_0^{+}(r_d,\omega) - \sum_i \mathbf{G}(r_i - r_d;\omega)\mathbf{D}_i(\omega)$$

Far field approximation $\mathbf{G}(r_i - r_d;\omega_0) \approx 3\mathbf{\Delta}_{r_d}^{\perp} e^{ik_0|\mathbf{r}_d - \mathbf{r}_i|} \approx 3\mathbf{\Delta}_{r_d}^{\perp} e^{ik_0 r_d(\hat{\mathbf{r}}_d - \hat{\mathbf{r}}_d\cdot\mathbf{r}_i)}$

$$I(\hat{\mathbf{r}}_d,t) = \left(\frac{3}{2k_0 r_d}\right)^2 \sum_{i,j} e^{ik_0(\mathbf{r}_i - \mathbf{r}_j)\cdot\hat{\mathbf{r}}_d}\text{Tr}_3[\mathbf{\Delta}_{r_d}^{\perp}\langle \mathbf{D}_i^{+}\otimes\mathbf{D}_j^{-}\rangle(t)]$$

We abandon the geometrical prefactor and compute the spectrum of the intensity

$$S(\hat{\mathbf{r}}_d,\omega) \;=\; 2\text{Re}\left\{\sum_{i,j}\sum_m s_{ij}^{m}(\hat{\mathbf{r}}_d)\langle D_{i,m}^{+}D_{j,m}^{-}\rangle(\omega)\right\}$$

where we introduced

- a prefactor coming from the interference of the light radiated by both dipoles

$$s_{ij}^{m}(\hat{\mathbf{r}}_d) = e^{ik_0(\mathbf{r}_i - \mathbf{r}_j)\cdot\hat{\mathbf{r}}_d}\mathbf{\Delta}_{r_d}^{\perp}\cdot\hat{\boldsymbol{e}}_m$$

- the spectrum of the correlators computed at a stationnary time t_s

$$\langle D_{i,m}^{+}D_{j,m}^{-}\rangle(\omega) = \int_0^{\infty} d\tau e^{-i\omega\tau}\langle D_{i,m}^{+}(t_s+\tau)D_{j,m}^{-}(t_s)\rangle$$

It is also interesting to compute the spectrum averaged over all directions

$$S(\omega) \;=\; \int_0^{\pi} d\theta \int_0^{2\pi} \sin\theta d\varphi S(\hat{\mathbf{r}}_d,\omega)$$

3 Atom pair

3.1 Atomic basis

3.1.1 Single polarization approximation

We consider a single polarization (non-degenerate two level system) in the previous equations of motion and can compute the full dynamics of the system by solving a closed set of 15 equations of motions. It is more convenient to introduce the population imbalance on atom i as $\Pi_i = [D_i^+, D_i^-] = \frac{1}{2}S_i^z$.

We solve

$$\frac{d}{dt}x = \mathcal{M}x + \lambda$$

where $x = (\langle D_1^+\rangle, \langle D_2^+\rangle, \langle D_1^-\rangle, \langle D_2^-\rangle, \langle D_1^- D_2^-\rangle, \langle D_1^+ D_2^-\rangle, \langle D_1^- D_2^+\rangle, \langle D_1^+ D_2^+\rangle, \langle \Pi_1\rangle, \langle \Pi_2\rangle, \langle \Pi_1\Pi_2\rangle, \langle D_1^+\Pi_2\rangle, \langle D_1^-\Pi_2\rangle, \langle \Pi_1 D_2^+\rangle, \langle \Pi_1 D_2^-\rangle)^T$

The coefficients of $\mathcal{M}$ come from

$$\frac{d}{dt}\langle D_1^+\rangle = -(i\Delta_p + 1/2)\langle D_1^+\rangle - \frac{i\Omega_1^-}{2}\langle \Pi_1\rangle + \frac{ig^*}{2}\langle \Pi_1 D_2^+\rangle$$

$$\frac{d}{dt}\langle D_2^+\rangle = -(i\Delta_p + 1/2)\langle D_2^+\rangle - \frac{i\Omega_2^-}{2}\langle \Pi_2\rangle + \frac{ig^*}{2}\langle D_1^+\Pi_2\rangle$$

$$\frac{d}{dt}\langle D_1^-\rangle = (i\Delta_p - 1/2)\langle D_1^-\rangle + \frac{i\Omega_1^+}{2}\langle \Pi_1\rangle - \frac{ig}{2}\langle \Pi_1 D_2^-\rangle$$

$$\frac{d}{dt}\langle D_2^-\rangle = (i\Delta_p - 1/2)\langle D_2^-\rangle + \frac{i\Omega_2^+}{2}\langle \Pi_2\rangle - \frac{ig}{2}\langle D_1^-\Pi_2\rangle$$

$$\frac{d}{dt}\langle D_1^- D_2^-\rangle = (2i\Delta_p - 1)\langle D_1^- D_2^-\rangle + \frac{i}{2}\Omega_1^+\langle \Pi_1 D_2^-\rangle + \frac{i}{2}\Omega_2^+\langle D_1^-\Pi_2\rangle$$

$$\frac{d}{dt}\langle D_1^- D_2^+\rangle = -\langle D_1^- D_2^+\rangle - \frac{ig}{4}\langle \Pi_1\rangle + \frac{ig^*}{4}\langle \Pi_2\rangle + \frac{i}{4}(g^* - g)\langle \Pi_1\Pi_2\rangle - \frac{i}{2}\Omega_2^-\langle D_1^-\Pi_2\rangle + \frac{i}{2}\Omega_1^+\langle \Pi_1 D_2^+\rangle$$

$$\frac{d}{dt}\langle D_1^+ D_2^-\rangle = -\langle D_1^+ D_2^-\rangle + \frac{ig^*}{4}\langle \Pi_1\rangle - \frac{ig}{4}\langle \Pi_2\rangle + \frac{i}{4}(g^* - g)\langle \Pi_1\Pi_2\rangle + \frac{i\Omega_2^+}{2}\langle D_1^+\Pi_2\rangle - \frac{i\Omega_1^-}{2}\langle \Pi_1 D_2^-\rangle$$

$$\frac{d}{dt}\langle D_1^+ D_2^+\rangle = -(2i\Delta_p + 1)\langle D_1^+ D_2^+\rangle - \frac{i\Omega_1^-}{2}\langle \Pi_1 D_2^+\rangle - \frac{i\Omega_2^-}{2}\langle D_1^+\Pi_2\rangle$$

$$\frac{d}{dt}\langle \Pi_1\Pi_2\rangle = i\,(g^* - g)\langle D_1^+ D_2^-\rangle + i\,(g^* - g)\langle D_1^- D_2^+\rangle - \langle \Pi_1\rangle - \langle \Pi_2\rangle - 2\langle \Pi_1\Pi_2\rangle - i\Omega_1^+\langle D_1^+\Pi_2\rangle + i\Omega_1^-\langle D_1^-\Pi_2\rangle - i\Omega_2^+\langle \Pi_1 D_2^+\rangle + i\Omega_2^-\langle \Pi_1 D_2^-\rangle$$

$$\frac{d}{dt}\langle \Pi_1\rangle = -1 + i\Omega_1^-\langle D_1^-\rangle - i\Omega_1^+\langle D_1^+\rangle - \langle \Pi_1\rangle + ig\langle D_1^+ D_2^-\rangle - ig^*\langle D_1^- D_2^+\rangle$$

$$\frac{d}{dt}\langle \Pi_2\rangle = -1 + i\Omega_2^-\langle D_2^-\rangle - i\Omega_2^+\langle D_2^+\rangle - \langle \Pi_2\rangle + ig\langle D_1^- D_2^+\rangle - ig^*\langle D_1^+ D_2^-\rangle$$

$$\frac{d}{dt}\langle D_1^+\Pi_2\rangle = -\langle D_1^+\rangle + \frac{ig}{2}\langle D_2^+\rangle + i\Omega_2^-\langle D_1^+ D_2^-\rangle - i\Omega_2^+\langle D_1^+ D_2^+\rangle - \frac{i}{2}\Omega_1^-\langle \Pi_1\Pi_2\rangle - (i\Delta_p + 3/2)\langle D_1^+\Pi_2\rangle + \frac{i\,(g - g^*)}{2}\langle \Pi_1 D_2^+\rangle$$

$$\frac{d}{dt}\langle D_1^-\Pi_2\rangle = -\langle D_1^-\rangle - \frac{ig^*}{2}\langle D_2^-\rangle - i\Omega_2^+\langle D_1^- D_2^+\rangle + i\Omega_2^-\langle D_1^- D_2^-\rangle + \frac{i}{2}\Omega_1^+\langle \Pi_1\Pi_2\rangle + (i\Delta_p - 3/2)\langle D_1^-\Pi_2\rangle +$$

$$\frac{i(g-g^*)}{2}\langle\Pi_1 D_2^-\rangle$$

$$\frac{d}{dt}\langle\Pi_1 D_2^+\rangle = \frac{ig}{2}\langle D_1^+\rangle - \langle D_2^+\rangle + i\Omega_1^-\langle D_1^- D_2^+\rangle - i\Omega_1^+\langle D_1^+ D_2^+\rangle - \frac{i}{2}\Omega_2^-\langle\Pi_1\Pi_2\rangle - (i\Delta_p + 3/2)\langle\Pi_1 D_2^+\rangle + \frac{i\,(g-g^*)}{2}\langle D_1^+\Pi_2\rangle$$

$$\frac{d}{dt}\langle\Pi_1 D_2^-\rangle = -\langle D_2^-\rangle - \frac{ig^*}{2}\langle D_1^-\rangle - i\Omega_1^+\langle D_1^+ D_2^-\rangle + i\Omega_1^-\langle D_1^- D_2^-\rangle + \frac{i}{2}\Omega_2^+\langle\Pi_1\Pi_2\rangle + (i\Delta_p - 3/2)\langle\Pi_1 D_2^-\rangle + \frac{i(g-g^*)}{2}\langle D_1^-\Pi_2\rangle$$

3.1.2 Three polarizations

With the quantization axis z, in the basis of the transitions $|g\rangle \to |e, m \equiv m_z\rangle$, the Green's function is

$$\mathbf{G}(r_{12}) = \frac{3}{2}\frac{e^{ikr_{12}}}{kr_{12}}\begin{pmatrix} \frac{1}{\sqrt{2}} & \frac{i}{\sqrt{2}} & 0 \\ 0 & 0 & 1 \\ \frac{-1}{\sqrt{2}} & \frac{i}{\sqrt{2}} & 0 \end{pmatrix}\begin{pmatrix} P-Q & 0 & 0 \\ 0 & P & 0 \\ 0 & 0 & P \end{pmatrix}\begin{pmatrix} \frac{1}{\sqrt{2}} & 0 & \frac{-1}{\sqrt{2}} \\ \frac{i}{\sqrt{2}} & 0 & \frac{-i}{\sqrt{2}} \\ 0 & 1 & 0 \end{pmatrix} = \frac{3}{2}\frac{e^{ikr_{12}}}{kr_{12}}\begin{pmatrix} \frac{2P-Q}{2} & 0 & \frac{Q}{2} \\ 0 & P & 0 \\ \frac{Q}{2} & 0 & \frac{2P-Q}{2} \end{pmatrix},$$

with $P \equiv P(ikr_{12})$ and $Q \equiv Q(ikr_{12})$. We now consider all three polarizations in the dynamics. The equations of motions can still be reduced to a set of 15 equations in a compact form,

$$\frac{d}{dt}\langle D_{1,m}^+\rangle = -(i\Delta_p + 1/2)\langle D_{1,m}^+\rangle + \frac{i}{2}\sum_{n,p}\left[G_{mn}^*\langle D_{1,p}^+ D_{1,p}^- D_{2,n}^+\rangle + G_{pn}^*\langle D_{1,m}^+ D_{1,p}^- D_{2,n}^+\rangle\right] - \frac{i}{2}\sum_n G_{mn}^*\langle D_{2,n}^+\rangle - \frac{i}{2}\sum_p\left[\Omega_{1p}^-\langle D_{1,m}^+ D_{1,p}^-\rangle + \Omega_{1m}^-\langle D_{1,p}^+ D_{1,p}^-\rangle\right] + \frac{i\Omega_{1m}^-}{2}$$

$$\frac{d}{dt}\langle D_{2,m}^+\rangle = -(i\Delta_p + 1/2)\langle D_{2,m}^+\rangle + \frac{i}{2}\sum_{n,p}\left[G_{mn}^*\langle D_{1,n}^+ D_{2,p}^+ D_{2,p}^-\rangle + G_{pn}^*\langle D_{1,n}^+ D_{2,m}^+ D_{2,p}^-\rangle\right] - \frac{i}{2}\sum_n G_{mn}^*\langle D_{1,n}^+\rangle - \frac{i}{2}\sum_p\left[\Omega_{2p}^-\langle D_{2,m}^+ D_{2,p}^-\rangle + \Omega_{2m}^-\langle D_{2,p}^+ D_{2,p}^-\rangle\right] + \frac{i\Omega_{2m}^-}{2}$$

$$\frac{d}{dt}\langle D_{1,m}^-\rangle = (i\Delta_p - 1/2)\langle D_{1,m}^+\rangle - \frac{i}{2}\sum_{n,p}\left[G_{mn}\langle D_{1,p}^+ D_{1,p}^- D_{2,n}^-\rangle + G_{pn}\langle D_{1,p}^+ D_{1,m}^- D_{2,n}^-\rangle\right] + \frac{i}{2}\sum_n G_{mn}\langle D_{2,n}^-\rangle + \frac{i}{2}\sum_p\left[\Omega_{1p}^+\langle D_{1,p}^+ D_{1,m}^-\rangle + \Omega_{1m}^+\langle D_{1,p}^+ D_{1,p}^-\rangle\right] - \frac{i\Omega_1^+}{2}$$

$$\frac{d}{dt}\langle D_{2,m}^-\rangle = (i\Delta_p - 1/2)\langle D_{2,m}^+\rangle - \frac{i}{2}\sum_{n,p}\left[G_{mn}\langle D_{1,n}^- D_{2,p}^+ D_{2,p}^-\rangle + G_{pn}\langle D_{1,n}^- D_{2,p}^+ D_{2,m}^-\rangle\right] + \frac{i}{2}\sum_n G_{mn}\langle D_{1,n}^-\rangle + \frac{i}{2}\sum_p\left[\Omega_{2p}^+\langle D_{2,p}^+ D_{2,m}^-\rangle + \Omega_{2m}^+\langle D_{2,p}^+ D_{2,p}^-\rangle\right] - \frac{i\Omega_2^+}{2}$$

$$\frac{d}{dt}\langle D_{1m}^- D_{2m'}^-\rangle = (2i\Delta_p - 1)\langle D_{1m}^- D_{2m'}^-\rangle + \frac{i}{2}\sum_p\left(\Omega_{1p}^+\langle D_{1p}^+ D_{1m}^- D_{2m'}^-\rangle + \Omega_{1m}^+\langle D_{1p}^+ D_{1p}^- D_{2m'}^-\rangle + \Omega_{2p}^+\langle D_{1m}^- D_{2p}^+ D_{2m'}^-\rangle + \Omega_{2m'}^+\langle D_{1m}^- D_{2p}^+ D_{2p}^-\rangle\right) - \frac{i}{2}(\Omega_{1m}^+\langle D_{2m'}^-\rangle + \Omega_{2m'}^+\langle D_{1m}^-\rangle)$$

$$\frac{d}{dt}\langle D_{1m}^+ D_{1m'}^- D_{2n}^-\rangle = (i\Delta_p - 3/2)\langle D_{1m}^+ D_{1m'}^- D_{2n}^-\rangle - \frac{i}{2}\sum_{n'} G_{mn'}^*\langle D_{1m}^+ D_{2n'}^+ D_{2n}^-\rangle + \frac{i}{2}\sum_p\left(\Omega_{2p}^+\langle D_{1m}^+ D_{1m'}^- D_{2p}^+ D_{2n}^-\rangle + \Omega_{2n}\langle D_{1m}^+ D_{1m'}^- D_{2p}^+ D_{2p}^-\rangle\right) + \frac{i}{2}(\Omega_{1m}^-\langle D_{1m'}^- D_{2n}^-\rangle - \Omega_{1m'}^+\langle D_{1m}^+ D_{2n}^-\rangle -$$

$\Omega^+_{2n}\langle D^+_{1m}D^-_{1m'}\rangle)$

etc until

$$\frac{d}{dt}\langle D^+_{1m}D^-_{1m'}D^+_{2n}D^-_{2n'}\rangle = -2\langle D^+_{1m}D^-_{1m'}D^+_{2n}D^-_{2n'}\rangle + \frac{i}{2}\Omega^-_{1m}\langle D^-_{1m'}D^+_{2n}D^-_{2n'}\rangle - \frac{i}{2}\Omega^+_{1m'}\langle D^+_{1m}D^+_{2n}D^-_{2n'}\rangle +$$
$$\frac{i}{2}\Omega^-_{2n}\langle D^+_{1m}D^-_{1m'}D^-_{2n'}\rangle - \frac{i}{2}\Omega^+_{2n'}\langle D^+_{1m}D^-_{1m'}D^+_{2n}\rangle + (1-\delta_{m'n})(G_{m'n}\langle D^+_{1m}D^-_{2n'}\rangle - G^*_{m'n}\langle D^+_{1m}D^-_{2n'}\rangle)$$

4 Equations of motion in the collective basis

4.1 Equation of motions

Single polarization approximation to start with. We now diagonalize in terms of collective modes. G can be diagonalized in a bi-orthogonal set of right and left eigenvectors. Because G is symmetric, the dual set of left and right eigenvectors are simply complex conjugate one of another. We call these eigenmodes *collective modes* because they diagonalize the full problem restricted to the sector with one excitation.

$$G_{ij} = \sum_k \Lambda_k \Psi_k(i)\Psi^*_k(j) = \sum_k \Lambda_k \Psi^*_k(i)\Psi_k(j)$$

with

$$G\Psi_n = \Lambda_n \Psi_n$$

$$\Lambda_n = -2(\omega_n - \omega_0)/\Gamma_0 + i\Gamma_n/\Gamma_0$$

We assume the basis is also orthonormal (numerically wrong, this is less true for denser systems) in order to be able to do unitary transformations using

$$S = \begin{pmatrix} \Psi_1(r_1) & \dots & \Psi_N(r_1) \\ \\ \Psi_1(r_N) & \dots & \Psi_N(r_N) \end{pmatrix}$$

$$\vec{\tilde{D}}^+ = S^\dagger \vec{D}^+$$

$$\vec{\tilde{\Omega}}^-_p = S^\dagger \vec{\Omega}^-_p$$

$$\tilde{\Pi} = S^\dagger \begin{pmatrix} \Pi_1 & & \\ & \ddots & \\ & & \Pi_N \end{pmatrix} S$$

$S^\dagger S = \mathbb{1} \Rightarrow \sum_i \Psi^*_n(i)\Psi_m(i) = \delta_{nm}$ and $SS^\dagger = \mathbb{1} \Rightarrow \sum_n \Psi_n(i)\Psi^*_n(j) = \delta_{ij}$.

The elements of $\tilde{\Pi}$ correspond to the overlap of different eigenfunctions, weighted by the local atomic populations.

$$\tilde{\Pi}_{nk} = \sum_{i=1}^{N} \Pi_i \Psi^*_n(i)\Psi_k(i) = (\Psi_n|\Psi_k)_\Pi$$

$G_{ij} = \sum_k \Lambda_k \Psi_k(i)\Psi^*_k(j) = \sum_k \Lambda_k \Psi^*_k(i)\Psi_k(j)$
$\tilde{D}^+_k = \sum_i \Psi^*_k(i) D^+_i$ and $D^+_i = \sum_k \Psi_k(i)\tilde{D}^+_k$
$\tilde{\Omega}^-_k = \sum_i \Psi^*_k(i)\Omega^-_i$ and $\Omega^-_i = \sum_k \Psi_k(i)\tilde{\Omega}^-_k$
$\tilde{\mathcal{F}}^\Pi_{nm} = \sum_i \Psi^*_n(i)\Psi_m(i)\mathcal{F}^\Pi_i$, $\tilde{\mathcal{F}}^+_n = \sum_i \Psi^*_k(i)\mathcal{F}^+_i$

Now, in this new basis, the dynamics of the dipole become

$$\begin{aligned}\frac{d}{dt}\tilde{D}_n^+ &= \sum_i \Psi_n^*(i)\left(-i\Delta_p D_i^+ + \frac{i}{2}\sum_j G_{ij}^* D_j^+ \Pi_i - \frac{i}{2}\Omega_i^- \Pi_i + \mathcal{F}_i^+\right) \\ &\quad -i\Delta_p \tilde{D}_n^+ + \frac{i}{2}\sum_{ijkh} \Lambda_k^* \Psi_n^*(i)\Psi_k(i)\Psi_k^*(j)\Psi_h(j)\tilde{D}_h^+\Pi_i - \frac{i}{2}\Omega_i^-\Pi_i + \tilde{\mathcal{F}}_n^+ \\ &= -i\Delta_p\tilde{D}_n^+ + \frac{i}{2}\sum_k \Lambda_k^*\tilde{D}_k^+ \sum_i \Psi_n^*(i)\Psi_k(i)\Pi_i - \frac{i}{2}\Omega_i^-\Pi_i + \tilde{\mathcal{F}}_n^+\end{aligned}$$

$$\frac{d}{dt}\tilde{D}_n^+ = -i\Delta_p\tilde{D}_n^+ + \frac{i}{2}\sum_k \Lambda_k^*\tilde{D}_k^+\tilde{\Pi}_{nk} - \frac{i}{2}\sum_k \tilde{\Omega}_{pk}^-\tilde{\Pi}_{nk} + \tilde{\mathcal{F}}_n^+$$

And the dynamics of the population imbalance

Using

$$\frac{d}{dt}\Pi_i = i\sum_j (D_i^+ G_{ij} D_j - D_j^+ G_{ij}^* D_i) - i(\Omega_{pi}^- D_i - D_i^+\Omega_i^+) + \mathcal{F}_i^\Pi$$

$G_{ij} = \sum_k \Lambda_k \Psi_k(i)\Psi_k^*(j) = \sum_k \Lambda_k \Psi_k^*(i)\Psi_k(j)$

$\tilde{D}_k^+ = \sum_i \Psi_k^*(i) D_i^+$ and $D_i^+ = \sum_k \Psi_k(i)\tilde{D}_k^+$

$\tilde{\Omega}_k^- = \sum_i \Psi_k^*(i)\Omega_i^-$ and $\Omega_i^- = \sum_k \Psi_k(i)\tilde{\Omega}_k^-$

$\tilde{\mathcal{F}}_{nm}^\Pi = \sum_i \Psi_n^*(i)\Psi_m(i)\mathcal{F}_i^\Pi$

$$\begin{aligned}\frac{d}{dt}\tilde{\Pi}_{nm} &= i\sum_{ijk} \Psi_n^*(i)\Psi_m(i)[D_i^+\Lambda_k\Psi_k^*(i)\Psi_k(j)D_j - D_j^+\Lambda_k^*\Psi_k(i)\Psi_k^*(j)D_i] - \\ &\quad i\sum_i [(\Psi_n^*(i)\Omega_i^-)(D_i\Psi_m(i)) - (\Psi_n^*(i)D_i^+)(\Omega_i^+\Psi_m(i))] + \tilde{\mathcal{F}}_{nm}^\Pi \\ &= i\sum_{hl} A_{hl}^{nm}\tilde{D}_h^+\tilde{D}_l^- + B_{hl}^{nm}(\tilde{D}_h^+\tilde{\Omega}_l^+ - \tilde{\Omega}_h^-\tilde{D}_l^-) + \tilde{\mathcal{F}}_{nm}^\Pi\end{aligned}$$

$$B_{hl}^{nm} = \sum_i \Psi_n^*(i)\Psi_m(i)\Psi_h(i)\Psi_l^*(i)$$

$$\begin{aligned}A_{hl}^{nm} &= \sum_k \left\{\Lambda_k \left(\sum_i \Psi_n^*(i)\Psi_m(i)\Psi_h(i)\Psi_k^*(i)\right)\left(\sum_j \Psi_k(j)\Psi_l^*(j)\right) - \right. \\ &\quad \left.\Lambda_k^*\left(\sum_i \Psi_n^*(i)\Psi_m(i)\Psi_k(i)\Psi_l^*(i)\right)\left(\sum_j \Psi_h(j)\Psi_k^*(j)\right)\right\} \\ &= \sum_k \left\{\Lambda_k B_{hk}^{nm}\left(\sum_j \Psi_k(j)\Psi_l^*(j)\right) - \Lambda_k^* B_{kl}^{nm}\left(\sum_j \Psi_h(j)\Psi_k^*(j)\right)\right\}\end{aligned}$$

Using the orthogonality condition over the sum on j,

$$A_{hl}^{nm} = B_{hl}^{nm}(\Lambda_l - \Lambda_h^*)$$

Finally

$$\frac{d}{dt}\tilde{D}_n^+ = -i\Delta_p\tilde{D}_n^+ + \frac{i}{2}\sum_k \Lambda_k^*\tilde{D}_k^+\tilde{\Pi}_{nk} - \frac{i}{2}\sum_k \tilde{\Omega}_k^-\tilde{\Pi}_{nk} + \tilde{\mathcal{F}}_n^+$$

$$\frac{d}{dt}\tilde{\Pi}_{nm} = i\sum_{hl} B_{hl}^{nm}\{(\Lambda_l - \Lambda_h^*)\tilde{D}_h^+\tilde{D}_l^- + (\tilde{D}_h^+\tilde{\Omega}_l^+ - \tilde{\Omega}_h^-\tilde{D}_l^-)\} + \tilde{\mathcal{F}}_{nm}^{\Pi}$$

$$B_{hl}^{nm} = \sum_i \Psi_n^*(i)\Psi_m(i)\Psi_h(i)\Psi_l^*(i)$$

We can note that $B_{nn}^{nn} = \mathrm{IPR}_n$!

4.2 Difference with atomic operators

For atomic operators, $(D_i^+)^2 = 0$ whereas for N_{at} atoms, $(\tilde{D}_n^+)^k \neq 0$ for $k \leqslant N_{\mathrm{at}}$. Therefore, even if we assume only one mode is populated and restrict the calculation to a single mode s, to get a closed system of equations for the dynamics, the equation of motion for terms of the form $(\tilde{D}_s^+)^n(\tilde{D}_s^-)^n$ for $n \leqslant N_{\mathrm{at}}$ are still required and a priori not negligeable with respect to RWA. I have no idea if truncating the hierachy of equations makes any sense.