

# 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  $\mu_d$ , 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  $\{\mathbf{k}, \boldsymbol{\epsilon}\}$ . We consider an addition external coherent pump at a drive frequency  $\omega_d$  and modeled by a classical field

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

The Hamiltonian of the complete system is

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

where we introduced a contact Hamiltonian  $H_c = \sum_{i \neq j} \mathbf{D}_i \cdot \mathbf{D}_j \delta(\mathbf{r}_i - \mathbf{r}_j) / \epsilon_0$  used to regularize later the divergence of the Green's function that we introduce in  $\mathbf{E}_\perp$

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

with the decay rate into the vacuum modes of the field

$$\Gamma_0 = \frac{\mu_d^2 k_0^3}{3\pi\epsilon_0\hbar},$$

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

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

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

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  $[\mathbf{D}_i^\pm, \mathbf{E}_\perp^\pm(\mathbf{r}_i)] = 0$ .

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

where we defined

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

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

- 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  $\mathbf{r} = \mathbf{0}$ )

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

One can finally obtain in the frame of the drive and with time and detuning  $\Delta$  in units of  $\Gamma_0^{-1}$ ,

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

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 explicitly, 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 Atom pair

### 2.1 Atomic basis

#### 2.1.1 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. This yields the correlators required to compute the fluorescence spectrum. It is more convenient to introduce the population imbalance on atom  $i$  as  $\Pi_i = [D_i^+, D_i^-] = \frac{1}{2}S_i^z$  and note that due to the closure relation  $\Pi_i = 2D_i^+ D_i^- - \mathbb{1}$ .

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

$$\begin{aligned}
\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 + \\
&\quad 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 + \\
&\quad \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 + \\
&\quad \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 + \\
&\quad \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 + \\
&\quad \frac{i(g - g^*)}{2}\langle D_1^- \Pi_2 \rangle
\end{aligned}$$

### 2.1.2 Three polarizations

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

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

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,

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

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

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

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

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

$$\Omega_{2p}^+ \langle D_{1m}^- D_{2p}^+ D_{2m}^- \rangle + \Omega_{2m}^+ \langle D_{1m}^- D_{2p}^+ D_{2p}^- \rangle) - \frac{i}{2} (\Omega_{1m}^+ \langle D_{2m}^- \rangle + \Omega_{2m}^+ \langle D_{1m}^- \rangle)$$

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

etc until

$$\begin{aligned} \frac{d}{dt} \langle D_{1m}^+ D_{1m}^- D_{2n}^+ D_{2n}^- \rangle &= -2 \langle D_{1m}^+ D_{1m}^- D_{2n}^+ D_{2n}^- \rangle + \frac{i}{2} \Omega_{1m}^- \langle D_{1m}^- D_{2n}^+ D_{2n}^- \rangle - \frac{i}{2} \Omega_{1m}^+ \langle D_{1m}^+ D_{2n}^+ D_{2n}^- \rangle + \\ &\quad \frac{i}{2} \Omega_{2n}^- \langle D_{1m}^+ D_{1m}^- D_{2n}^- \rangle - \frac{i}{2} \Omega_{2n}^+ \langle D_{1m}^+ D_{1m}^- D_{2n}^+ \rangle + (1 - \delta_{m'n}) (G_{m'n} \langle D_{1m}^+ D_{2n}^- \rangle - G_{m'n}^* \langle D_{1m}^+ D_{2n}^- \rangle) \end{aligned}$$