Populations rate equations for an atom with an hyperfine structure

Abstract

Some notes on the derivation of rate equations for ground and excited populations, disregarding coherences, from the master equation. Based on [SteckQAO, §7.8].

1. The population rate equations

We consider a transition between two levels J_g (ground) and J_e (excited) which are hyperfine-split. We want to know the population transfer rate between the different states. From the master equation (ie. optical Bloch equations), under the approximation that the optical coherences are static and that there are no other coherences, we can obtain the following population rate equations :

$$\begin{aligned}
\partial_t P_g &= \sum_e \left(+\mathcal{F}_{eg}\,\delta_{q,q_{\mathsf{field}}}\,R(\mathcal{I})\,\mathcal{L}(\Delta_{eg})\,(P_e - P_g) + \mathcal{F}_{eg}\,\Gamma\,P_e\right)\\
\partial_t P_e &= \sum_g \left(-\mathcal{F}_{eg}\,\delta_{q,q_{\mathsf{field}}}\,R(\mathcal{I})\,\mathcal{L}(\Delta_{eg})\,(P_e - P_g)\right) - \Gamma\,P_e
\end{aligned}$$

with

$$\begin{aligned}
\mathcal{L}(\Delta) &= \frac{1}{1+(2\,\Delta/\Gamma)^2} \text{ the lorentzian lineshape}\\
\mathcal{F}_{eg} &= (2\,F_e+1)\,(2\,J_e+1)\left\{\begin{matrix} J_g & J_e & 1\\ F_e & F_g & I\end{matrix}\right\}^2 (2\,F_g+1)\begin{pmatrix} F_e & 1 & F_g\\ M_e & q & -M_g\end{pmatrix}^2,\ q = M_g - M_e\\
R(\mathcal{I}) &= \frac{6\pi\,c^2\,\mathcal{I}}{\hbar\,\omega_0^3} = \frac{\Gamma}{2}\frac{\mathcal{I}}{\mathcal{I}_{\mathsf{sat},0}}, \text{ where } \mathcal{I} \text{ is the intensity}
\end{aligned}$$

P_i is the population in the state $i=(F,M_F)$; labels $i=g,e$ refer to the ground and excited states respectively. The sums $\sum_i = \sum_{F,M_F}$ are over possible angular momentum states given J_i and the nuclear spin I. The populations are normalized such that $\sum_g P_g + \sum_e P_e = 1$.

Importantly, we assume that the light field has a **single polarization component** $q = q_{\mathsf{field}}$ in the chosen quantization axis[1]. While these rate equations are often still valid even in the presence of multiple components (such as $\sigma^+ + \sigma^-$) in the presence of Zeeman splitting, they are certainly wrong in the absence of Zeeman splitting. Indeed, with multiple components, we must in general keep track of the coherences between M_e states and between M_g states to represent the field-induced orientation of the atom, as discussed in [SteckQAO, §7.8.3.1, Single Field Polarization], and we cannot have a closed system for populations only. Additionally, we ignore any F_e-F_e' and F_g-F_g' coherences, as discussed in [SteckQAO, §7.8.3.3]. In the presence of multiple optical fields, one can add them incoherently (ie. by simply adding terms $\delta_{q,q_{\mathsf{field},k}}\,R(\mathcal{I}_k)\,\mathcal{L}(\Delta_{eg,k})\,(P_e - P_g)$) if the non-linear coherent effects can be ignored, as discussed in [SteckQAO, §7.8.3.2, Multiple Fields].

$\hbar\,\Delta_{eg} = \hbar\,\Delta_{\mathsf{bary}} + \Delta E^g(F_g,M_g) - \Delta E^e(F_e,M_e)$ is the detuning from (F_g,M_g) to (F_e,M_e), with ΔE^g and ΔE^e the hyperfine and Zeeman energy shifts in the ground and excited manifolds, and Δ_{bary} the detuning of the light field between the barycenters (non-split) manifolds.

Here, $\Gamma = \Gamma_{J_g\text{-}J_e}$ is the inverse lifetime at the *fine-structure* level. $\mathcal{I}_{\mathsf{sat}}$ is the standard saturation intensity at the fine structure level, $\mathcal{I}_{\mathsf{sat},0} = \frac{\hbar\,\omega_0^3\,\Gamma}{12\,\pi\,c^2}$, where $\hbar\omega_0$ is the J_g-J_e energy difference. $\left\{\begin{smallmatrix} \cdot & \cdot & \cdot\\ \cdot & \cdot & \cdot\end{smallmatrix}\right\}$ and $\left(\begin{smallmatrix} \cdot & \cdot & \cdot\\ \cdot & \cdot & \cdot\end{smallmatrix}\right)$ are respectively the 6j-symbols and 3j-symbols, and $\mathcal{F}_{eg}$ can also be written with Clebsch-Gordan coefficients : $\mathcal{F}_{eg} = (2\,F_e+1)\,(2\,J_e+1)\left\{\begin{smallmatrix} J_g & J_e & 1\\ F_e & F_g & I\end{smallmatrix}\right\}^2 \langle F_e, M_e; 1, q | F_g, M_g\rangle^2$. We see that the decay rate of any excited state e is Γ, with branching ratios $\mathcal{F}_{eg}$ to ground states g. Naturally, we have that $\sum_g \mathcal{F}_{eg} = 1$ (eq. 3.3).

1. ie. if we decompose the field as $\vec{\mathcal{E}}(t) = \sum_q (-1)^q\,\vec{e}_{-q}\,\mathcal{E}_{0q}^{(+)}\,\mathrm{e}^{\mathrm{i}\omega t} + \mathrm{c.c.}$, we have a single non-zero $\mathcal{E}_{0q}^{(+)}$ for $q = q_{\mathsf{field}}$.

2. Main steps of the derivation

We use the framework and conventions found in Steck's Quantum and Atom Optics notes [SteckQAO]. We start from the master equation when driving a $J_g \longleftrightarrow J_e$ transition [SteckQAO, (7.512)] :

$$\hbar\,\partial_t\,\tilde{\rho} \;=\; -\mathrm{i}\left[\tilde{\boldsymbol{H}}_{\text{atom}},\tilde{\rho}\right]-\mathrm{i}\left[\tilde{\boldsymbol{H}}_{\text{atom-light}},\tilde{\rho}\right]+\sum_q \hbar\,\Gamma\,\frac{2\,J_e+1}{2\,J_g+1}\,\mathcal{D}[\boldsymbol{\Sigma}_q,\tilde{\rho}]$$

where

$$\begin{aligned}
\tilde{\boldsymbol{H}}_{\text{atom}} &= \sum_e\left(\Delta E^e(e)-\hbar\,\Delta\right)|e\rangle\langle e|+\sum_g \Delta E^g(g)\,|g\rangle\langle g|\\
\tilde{\boldsymbol{H}}_{\text{atom-light}} &= \frac{\hbar}{2}\sum_q\left(\Omega_q^*\,\boldsymbol{\Sigma}_q+\Omega_q\,\boldsymbol{\Sigma}_q^\dagger\right),\quad \Omega_q=-\frac{2}{\hbar}\,\langle J_g\|\boldsymbol{d}\|J_e\rangle_{\text{Steck}}\,\mathcal{E}_{0q}^{(+)}\\
\mathcal{D}[\boldsymbol{\Sigma},\rho] &= \boldsymbol{\Sigma}\,\rho\,\boldsymbol{\Sigma}^\dagger-\frac{1}{2}\left(\boldsymbol{\Sigma}^\dagger\,\boldsymbol{\Sigma}\,\rho+\rho\,\boldsymbol{\Sigma}^\dagger\,\boldsymbol{\Sigma}\right)\\
\boldsymbol{\Sigma}_q &= \sum_{g,e} f_{ge,q}\,|g\rangle\langle e|\quad \text{(lowering operators)}\\
f_{ge,q} &= (-1)^{F_e+J_g+1+I}\sqrt{2\,F_e+1}\,\sqrt{2\,J_g+1}\begin{Bmatrix} J_g & J_e & 1\\ F_e & F_g & I\end{Bmatrix}\langle F_g,M_g|F_e,M_e;1,q\rangle
\end{aligned}$$

where running g and e in sums correspond to ground and excited states, here (F_g,M_g) and (F_e,M_e). $\Delta E^e(F_e,M_e)$ and $\Delta E^g(F_g,M_g)$ correspond to the energy splitting of the states relative to the level barycenter (ie. hyperfine + Zeeman splittings).

If we neglect e-e' and g-g' coherences, ie. if $\tilde{\rho}_{gg'}=\delta_{gg'}\,\tilde{\rho}_{gg}$ and $\tilde{\rho}_{ee'}=\delta_{ee'}\,\tilde{\rho}_{ee}$, we obtain :

$$\begin{aligned}
\partial_t\,\tilde{\rho}_{gg} &= \langle g|\,\partial_t\,\tilde{\rho}\,|g\rangle\\
&= +\sum_q\sum_e\left(-\frac{\mathrm{i}}{2}\,f_{ge,q}\left(\Omega_q^*\,\tilde{\rho}_{ge}^*-\Omega_q\,\tilde{\rho}_{ge}\right)+\Gamma\,\frac{2\,J_e+1}{2\,J_g+1}\,f_{ge,q}^2\,\tilde{\rho}_{ee}\right) \qquad (1)\\
\partial_t\,\tilde{\rho}_{ee} &= \langle e|\,\partial_t\,\tilde{\rho}\,|e\rangle\\
&= -\sum_q\sum_g\left(-\frac{\mathrm{i}}{2}\,f_{ge,q}\left(\Omega_q^*\,\tilde{\rho}_{ge}^*-\Omega_q\,\tilde{\rho}_{ge}\right)+\Gamma\,\frac{2\,J_e+1}{2\,J_g+1}\,f_{ge,q}^2\,\tilde{\rho}_{ee}\right) \qquad (2)
\end{aligned}$$

To close the system, we need the optical coherences $\tilde{\rho}_{ge}$. From the master equation, we have

$$\begin{aligned}
\partial_t\,\tilde{\rho}_{ge} &= \langle g|\,\partial_t\,\tilde{\rho}\,|e\rangle\\
&= -\mathrm{i}\,\Delta_{eg}\,\tilde{\rho}_{ge}-\frac{\mathrm{i}}{2}\sum_q f_{ge,q}\,\Omega_q^*\left(\tilde{\rho}_{ee}-\tilde{\rho}_{gg}\right)-\frac{1}{2}\,\Gamma\,\tilde{\rho}_{ge} \qquad (3)
\end{aligned}$$

Under the adiabatic approximation (steady-state optical coherences), $\partial_t\,\tilde{\rho}_{ge}\approx 0$, and we thus obtain

$$\tilde{\rho}_{ge}\approx -\frac{\mathrm{i}}{2}\left(\tilde{\rho}_{ee}-\tilde{\rho}_{gg}\right)\frac{1}{\mathrm{i}\,\Delta_{eg}+\Gamma/2}\sum_q f_{ge,q}\,\Omega_q^*$$

Inserting these coherences in (1), we can now close the system and get[2]

$$\partial_t\,\tilde{\rho}_{gg} \;=\; \sum_e\left(\frac{1}{4}\left(\tilde{\rho}_{ee}-\tilde{\rho}_{gg}\right)f_{ge,q}^2\,|\Omega_q|^2\,\frac{\Gamma}{\Delta_{eg}^2+\Gamma^2/4}+\Gamma\,\frac{2\,J_e+1}{2\,J_g+1}\,f_{ge,q}^2\,\tilde{\rho}_{ee}\right)$$

2. Details : inserting the coherences in $\partial_t\,\tilde{\rho}_{gg}$, we get

$$\sum_e\left(\frac{1}{4}\left(\tilde{\rho}_{ee}-\tilde{\rho}_{gg}\right)\left(\frac{1}{-\mathrm{i}\Delta_{eg}+\frac{\Gamma}{2}}\sum_q\sum_{q'}\Omega_q^* f_{ge,q}f_{ge,q'}\Omega_{q'}+\frac{1}{\mathrm{i}\Delta_{eg}+\frac{\Gamma}{2}}\sum_q\sum_{q'}\Omega_q f_{ge,q}f_{ge,q'}\Omega_q^*\right)+\sum_q\Gamma\,\frac{2\,J_e+1}{2\,J_g+1}\,f_{ge}^2\,\tilde{\rho}_{ee}\right)$$

Because of the CB coeffs in $f_{ge,q}$'s, we must have $M_e+q=M_g=M_e+q'$, so $q=q'$ in the $\sum_q\sum_{q'}$ sums, which both simplify to $\sum_q f_{ge,q}^2\,\Omega_q^*\,\Omega_q$. Each (e,g) pair is actually associated with a q such that $M_e+q=M_g$, ie. there is a single term in the sum, that we write $f_{ge,q}^2\,|\Omega_q|^2$, where is assumed to be $q=M_g-M_e$. For the same reason, we now omit the $\sum_q$ sum in the decay term. $\partial_t\,\tilde{\rho}_{gg}$ then reads

$$\sum_e\left(\frac{1}{4}\left(\tilde{\rho}_{ee}-\tilde{\rho}_{gg}\right)f_{ge,q}^2\,|\Omega_q|^2\underbrace{\left(\frac{1}{-\mathrm{i}\,\Delta_{eg}+\frac{\Gamma}{2}}+\frac{1}{\mathrm{i}\,\Delta_{eg}+\frac{\Gamma}{2}}\right)}_{=\frac{\Gamma}{\Delta_{eg}^2+\Gamma^2/4}}+\Gamma\,\frac{2\,J_e+1}{2\,J_g+1}\,f_{ge}^2\,\tilde{\rho}_{ee}\right)$$

where, implicitly, $q = M_g - M_e$. The Rabi frequencies Ω_q have the nasty property of being dependant on the Wigner-Eckart convention used for the RDME. Let's use [SteckQAO, (7.305)]

$$\Gamma = \Gamma_{J_e\text{-}J_g} = \frac{\omega_0^3}{3\pi\,\epsilon_0\,\hbar\,c^3}\,\frac{2\,J_g+1}{2\,J_e+1}\,|\langle J_g\|\boldsymbol{d}\|J_e\rangle_{\text{Steck}}|^2$$

so that

$$|\Omega_q|^2 = \frac{4}{\hbar^2}\,\Gamma\,\frac{2\,J_e+1}{2\,J_g+1}\,\frac{3\pi\,\epsilon_0\,\hbar\,c^3}{\omega_0^3}\,|\mathcal{E}_{0q}^{(+)}|^2$$

Then,

$$\partial_t\,\tilde{\rho}_{gg} = \sum_e \mathcal{F}_{eg,q}\left((\tilde{\rho}_{ee}-\tilde{\rho}_{gg})\,R_q\,\frac{1}{4\,\Delta_{eg}^2/\Gamma^2+1}+\Gamma\,\tilde{\rho}_{ee}\right) \tag{4}$$

where we collected

$$\begin{aligned}\mathcal{F}_{eg,q} &= \frac{2\,J_e+1}{2\,J_g+1}\,f_{ge,q}^2 \\ &= \frac{2\,J_e+1}{\cancel{2\,J_g+1}}\,(-1)^{2\underbrace{(F_e+J_g+1+I)}_{\text{integer}}}\,(2\,F_e+1)\,\cancel{(2\,J_g+1)}\left\{\begin{array}{ccc}J_g & J_e & 1\\ F_e & F_g & I\end{array}\right\}^2\langle F_g,M_g|F_e,M_e;1,q\rangle^2 \\ &= (2\,F_e+1)\,(2\,J_e+1)\left\{\begin{array}{ccc}J_g & J_e & 1\\ F_e & F_g & I\end{array}\right\}^2(2\,F_g+1)\begin{pmatrix}F_e & 1 & F_g\\ M_e & q & -M_g\end{pmatrix}^2\end{aligned} \tag{5}$$

and absorbed the Γ coming from the RDME into the lorentzian lineshape. We also defined a coupling rate

$$R_q = \frac{3\pi\,\epsilon_0\,c^3}{\hbar\,\omega_0^3}\,4\,\left|\mathcal{E}_{0q}^{(+)}\right|^2 = \delta_{q,q_{\text{field}}}\,\frac{6\pi\,c^2}{\hbar\,\omega_0^3}\,\mathcal{I} \tag{6}$$

using the fact that the intensity is $\mathcal{I} = 2\,\epsilon_0\,c\,\left|\mathcal{E}_0^{(+)}\right|^2$. We assumed that only one field mode $q = q_{\text{field}}$ exists. Anyways, if it is not the case, we cannot use the above derivation, which discarded the coherences between M_F states. Indeed, a superposition of field modes will create a coherent superposition of population in M_F states, which cannot be assumed to be zero in that case. The rate R_q does not depend on the RDME/Γ. However, if we want to make appear the usual saturation intensity $\mathcal{I}_{\text{sat}}$, we can write it as

$$R_q = \delta_{q,q_{\text{field}}}\,\frac{\Gamma}{2}\,\frac{\mathcal{I}}{\mathcal{I}_{\text{sat}}}\quad\text{with}\quad\mathcal{I}_{\text{sat}} = \frac{\hbar\,\omega_0^3\,\Gamma}{12\,\pi\,c^2} \tag{7}$$

Sanity check : if we have a single transition $e \leftrightarrow g$ at resonance ($\Delta_{eg} = 0$), the steady state $\partial_t\,\tilde{\rho}_{gg} = 0$ is

$$(\tilde{\rho}_{ee}-\tilde{\rho}_{gg})\,R+\Gamma\,\tilde{\rho}_{ee} = 0 \quad \underset{\tilde{\rho}_{ee}+\tilde{\rho}_{gg}=1}{\Longleftrightarrow} \quad \tilde{\rho}_{ee} = \frac{R}{2\,R+\Gamma} = \frac{1}{2}\,\frac{s}{s+1}\ \text{with}\ s = \frac{\mathcal{I}}{\mathcal{I}_{\text{sat}}}$$

which is a familiar result.

Note that the resulting population rate equations can more easily be obtained from the system (7.535, hyperfine-structure rate equations) in [SteckQAO] by ignoring coherences (taking $\tilde{\rho}_{F_g M_g, F_g' M_g'} = \delta_{F_g F_g'}\,\delta_{M_g M_g'}\,P_g$, same for e)[3], but we wanted here to have an explicit and self-contained derivation.

3. Derivation details

3.1. $\partial_t\,\tilde{\rho}_{gg}$

Let us give some details and insights on the derivation of (1). We have

$$\begin{aligned}\hbar\,\partial_t\,\tilde{\rho}_{gg} &= \langle g|\,\hbar\,\partial_t\,\tilde{\rho}\,|g\rangle \\ &\quad -\mathrm{i}\,\langle g|[\tilde{\boldsymbol{H}}_{\text{atom}},\tilde{\rho}]|g\rangle-\mathrm{i}\,\langle g|[\tilde{\boldsymbol{H}}_{\text{atom-light}},\tilde{\rho}]|g\rangle+\sum_q\hbar\,\Gamma\,\frac{2\,J_e+1}{2\,J_g+1}\,\langle g|\mathcal{D}[\boldsymbol{\Sigma}_q,\tilde{\rho}]|g\rangle\end{aligned}$$

Atom term

3. Ignoring coherences, we easily obtain from (7.535) that $\partial_t\,\tilde{\rho}_{ee} = \sum_g |\Omega(g,e)|^2\,(\tilde{\rho}_{gg}-\tilde{\rho}_{ee})\,\frac{\Gamma^{-1}}{1+(2\,\Delta_{ge}/\Gamma)^2}-\Gamma\,\tilde{\rho}_{ee}$. Then, inserting $\Omega(g,e) = \Omega_q\,f_{ge,q}$ (7.422, sublevel Rabi frequencies) [SteckQAO], we obtain our population rate equations.

The first term is 0 because $\tilde{\boldsymbol{H}}_{\mathsf{atom}}$ has operators $|e'\rangle\langle e'|$ and $|g'\rangle\langle g'|$, and

- $\big\langle g\big|\big[|e'\rangle\langle e'|,\tilde{\rho}\big]\big|g\big\rangle = \underbrace{\langle g||e'\rangle}_{=0}\langle e'|\tilde{\rho}|g\rangle - \langle g|\tilde{\rho}|e'\rangle\underbrace{\langle e'||g\rangle}_{=0} = 0$

- $\big\langle g\big|\big[|g'\rangle\langle g'|,\tilde{\rho}\big]\big|g\big\rangle = \langle g||g'\rangle\langle g'|\tilde{\rho}|g\rangle - \langle g|\tilde{\rho}|g'\rangle\langle g'||g\rangle = \delta_{gg'}(\tilde{\rho}_{gg}-\tilde{\rho}_{gg}) = 0$

Atom-light term

Let's now tackle the atom-light interaction term. We use the definitions of $\tilde{\boldsymbol{H}}_{\mathsf{atom\text{-}light}}$ and $\boldsymbol{\Sigma}_q$ and develop:

$$\begin{aligned}\langle g|[\tilde{\boldsymbol{H}}_{\mathsf{atom\text{-}light}},\tilde{\rho}]|g\rangle &= \frac{\hbar}{2}\sum_q\left(\Omega_q^*\,\langle g|[\boldsymbol{\Sigma}_q,\tilde{\rho}]|g\rangle + \Omega_q\,\langle g|[\boldsymbol{\Sigma}_q^\dagger,\tilde{\rho}]|g\rangle\right)\\ &= \frac{\hbar}{2}\sum_q\sum_{g',e'} f_{g'e',q}\Big(\Omega_q^*\,\langle g|\big[|g'\rangle\langle e'|,\tilde{\rho}\big]|g\rangle + \Omega_q\,\langle g|\big[|e'\rangle\langle g'|,\tilde{\rho}\big]|g\rangle\Big)\end{aligned} \tag{8}$$

using that $(f_{ge,q}\,|g\rangle\langle e|)^\dagger = f_{ge,q}^*\,|e\rangle\langle g|$ and that $f_{ge,q}$'s are real numbers with the phase convention we use for Clebsch-Gordan coefficients. We then have

$$\langle g|\big[|g'\rangle\langle e'|,\tilde{\rho}\big]|g\rangle = \langle g||g'\rangle\langle e'|\tilde{\rho}|g\rangle - \langle g|\tilde{\rho}|g'\rangle\underbrace{\langle e'||g\rangle}_{=0} = \delta_{gg'}\,\tilde{\rho}_{e'g} - 0$$

$$\langle g|\big[|e'\rangle\langle g'|,\tilde{\rho}\big]|g\rangle = \overbrace{\langle g||e'\rangle}^{=0}\langle g'|\tilde{\rho}|g\rangle - \langle g|\tilde{\rho}|e'\rangle\langle g'||g\rangle = 0 - \delta_{gg'}\,\tilde{\rho}_{ge'}$$

Thus,

$$\langle g|[\tilde{\boldsymbol{H}}_{\mathsf{atom\text{-}light}},\tilde{\rho}]|g\rangle = \frac{\hbar}{2}\sum_q\sum_{e'} f_{ge',q}\Big(\Omega_q^*\,\tilde{\rho}_{ge'}^* - \Omega_q\,\tilde{\rho}_{ge'}\Big)$$

Decay term

The decay term takes longer to derive. We have to evaluate the $\langle g|\cdot|g\rangle$ element of $\mathcal{D}[\boldsymbol{\Sigma}_q,\tilde{\rho}] = \boldsymbol{\Sigma}_q\,\rho\,\boldsymbol{\Sigma}_q^\dagger - \frac{1}{2}\left(\boldsymbol{\Sigma}_q^\dagger\,\boldsymbol{\Sigma}_q\,\rho + \rho\,\boldsymbol{\Sigma}_q^\dagger\,\boldsymbol{\Sigma}_q\right)$. For each of these three terms, we have in the product two lowering or raising operator, and thus sums $\sum_{g'e'}$. Let's thus expand all three terms at once. For $\boldsymbol{\Sigma}_q$'s we have $|g'\rangle\langle e'|$, while for $\boldsymbol{\Sigma}_q^\dagger$'s we have $|e'\rangle\langle g'|$. We thus have

$$\begin{aligned}\langle g|\mathcal{D}[\boldsymbol{\Sigma}_q,\tilde{\rho}]|g\rangle &= \sum_{g',e'} f_{g'e',q}\sum_{g'',e''} f_{g''e'',q} \\ &\quad\Big(\langle g||g'\rangle\langle e'|\tilde{\rho}|e''\rangle\langle g''||g\rangle - \tfrac{1}{2}\big(\overbrace{\langle g||e''\rangle}^{=0}\langle g''||g'\rangle\langle e'|\tilde{\rho}|g\rangle + \langle g|\tilde{\rho}|e''\rangle\langle g''||g'\rangle\overbrace{\langle e'||g\rangle}^{=0}\big)\Big)\\ &= \sum_{g',e'} f_{g'e',q}\sum_{g'',e''} f_{g''e'',q}\,\delta_{gg'}\,\tilde{\rho}_{e'e''}\,\delta_{gg''} - \tfrac{1}{2}(0+0)\\ &= \sum_{e',e''} f_{ge',q}\,f_{ge'',q}\,\tilde{\rho}_{e'e''}\end{aligned} \tag{9}$$

It is a significant pain to carry all these e'-e'' coherences. Actually, this can be partially simplified away by noticing that the $f_{ge,q}$'s contain Clebsch-Gordan coefficients $\langle F_g,M_g|F_e,M_e;1,q\rangle$, which are non-zero only if $M_g = M_e + q$. The product $f_{ge',q}\,f_{ge'',q}$ can thus be non-zero only if $M_e' + q = M_g = M_e'' + q$, and thus $M_e' = M_e''$. Thus, the double sum simplifies as

$$\langle g|\mathcal{D}[\boldsymbol{\Sigma}_q,\tilde{\rho}]|g\rangle = \sum_{F_e',M_e',F_e''} f_{F_g,M_g,F_e',M_e',q}\,f_{F_g,M_g,F_e'',M_e',q}\,\tilde{\rho}_{F_e',M_e',F_e'',M_e'}$$

Still, in most cases the hyperfine coherences $\tilde{\rho}_{F_e',M_e',F_e'',M_e'}$ with $F_e' \neq F_e''$ can be neglected, in particular when the time-scales are large compared the the hyperfine splittings. We thus further simplify the problem by assuming that there is no coherence in-between hyperfine manifolds, as mentioned in the main text : $\tilde{\rho}_{F_e',M_e',F_e'',M_e'} \approx \delta_{F_e'F_e''}\,\tilde{\rho}_{e'e'}$. We then reduce the sum $\sum_{F_e',M_e',F_e''}$ to $\sum_{e'=(F_e',M_e')}$ and obtain

$$\langle g|\mathcal{D}[\boldsymbol{\Sigma}_q,\tilde{\rho}]|g\rangle \approx \sum_{e'} f_{ge',q}^2\,\tilde{\rho}_{e'e'}$$

Putting all terms together, we obtain (1) for $\partial_t \tilde{\rho}_{gg}$, which depend only on excited populations $\tilde{\rho}_{e'e'}$ and optical coherences $\tilde{\rho}_{ge'}$, in the approximation we made.

3.2. $\partial_t \tilde{\rho}_{ge}$

Let us give some details and insights on the derivation of (3). We have

$$\hbar\,\partial_t\,\tilde{\rho}_{ge} \;=\; -\mathrm{i}\,\langle g|[\tilde{\boldsymbol{H}}_{\text{atom}},\tilde{\rho}]|e\rangle - \mathrm{i}\,\langle g|[\tilde{\boldsymbol{H}}_{\text{atom-light}},\tilde{\rho}]|e\rangle + \sum_q \hbar\,\Gamma\,\tfrac{2\,J_e+1}{2\,J_g+1}\,\langle g|\mathcal{D}[\boldsymbol{\Sigma}_q,\tilde{\rho}]|e\rangle$$

Atom term

For the first term, it is easy to get

$$\begin{aligned}\langle g|[\tilde{\boldsymbol{H}}_{\text{atom}},\tilde{\rho}]|e\rangle \;&=\; \big(-\Delta E^e(e)+\hbar\,\Delta+\Delta E^g(g)\big)\,\tilde{\rho}_{ge}\\ &=:\; \hbar\,\Delta_{eg}\,\tilde{\rho}_{ge}\end{aligned}$$

Atom-light term

From (3.1.2), we just change the second g by e :

$$\langle g|[\tilde{\boldsymbol{H}}_{\text{atom-light}},\tilde{\rho}]|e\rangle \;=\; \frac{\hbar}{2}\sum_q\sum_{g',e'} f_{g'e',q}\Big(\Omega_q^*\,\langle g|\big[|g'\rangle\langle e'|,\tilde{\rho}\big]|e\rangle + \Omega_q\,\langle g|\big[|e'\rangle\langle g'|,\tilde{\rho}\big]|e\rangle\Big)$$

Then,

$$\begin{aligned}\langle g|\big[|g'\rangle\langle e'|,\tilde{\rho}\big]|e\rangle \;&=\; \langle g||g'\rangle\langle e'|\tilde{\rho}|e\rangle - \langle g|\tilde{\rho}|g'\rangle\langle e'||e\rangle \;=\; \delta_{gg'}\,\tilde{\rho}_{e'e}-\delta_{ee'}\,\tilde{\rho}_{gg'}\\ \langle g|\big[|e'\rangle\langle g'|,\tilde{\rho}\big]|e\rangle \;&=\; \underbrace{\langle g||e'\rangle}_{=0}\langle g'|\tilde{\rho}|e\rangle - \langle g|\tilde{\rho}|e'\rangle\underbrace{\langle g'||e\rangle}_{=0} \;=\; 0\end{aligned}$$

We thus obtain

$$\langle g|[\tilde{\boldsymbol{H}}_{\text{atom-light}},\tilde{\rho}]|e\rangle \;=\; \frac{\hbar}{2}\sum_q \Omega_q^*\left(\sum_{e'} f_{ge',q}\,\tilde{\rho}_{e'e} - \sum_{g'} f_{g'e,q}\,\tilde{\rho}_{gg'}\right) \tag{10}$$

Here, we cannot drop the M_F coherences between excited states and between ground states in general. If we still insist that $\tilde{\rho}_{e'e}\approx\delta_{ee'}\,\tilde{\rho}_{ee}$ and that $\tilde{\rho}_{gg'}\approx\delta_{gg'}\,\tilde{\rho}_{gg}$, we obtain a familiar flipping term

$$\langle g|[\tilde{\boldsymbol{H}}_{\text{atom-light}},\tilde{\rho}]|e\rangle \;\approx\; \left(\frac{\hbar}{2}\sum_q \Omega_q^*\,f_{ge,q}\right)(\tilde{\rho}_{ee}-\tilde{\rho}_{gg}) \tag{11}$$

Decay term

The decay term is more tricky to handle. Again, re-using (9) and replacing the second g by e, we have

$$\begin{aligned}\langle g|\mathcal{D}[\boldsymbol{\Sigma}_q,\tilde{\rho}]|e\rangle \;&=\; \sum_{g',e'} f_{g'e',q}\sum_{g'',e''} f_{g''e'',q} \\ &\quad\Big(\langle g||g'\rangle\langle e'|\tilde{\rho}|e''\rangle\overbrace{\langle g''||e\rangle}^{=0} - \tfrac{1}{2}\big(\overbrace{\langle g||e''\rangle}^{=0}\langle g''||g'\rangle\langle e'|\tilde{\rho}|e\rangle + \langle g|\tilde{\rho}|e''\rangle\langle g''||g'\rangle\langle e'||e\rangle\big)\Big)\\ &=\; -\frac{1}{2}\sum_{g',e'} f_{g'e',q}\sum_{g'',e''} f_{g''e'',q}\,\tilde{\rho}_{ge''}\,\delta_{g'g''}\,\delta_{e'e}\\ &=\; -\frac{1}{2}\sum_{e'',g''} f_{g''e,q}\,f_{g''e'',q}\,\tilde{\rho}_{ge''}\end{aligned} \tag{12}$$

One should't try to simplify that directly. The trick is to consider the sum over q,

$$\sum_q \langle g|\mathcal{D}[\boldsymbol{\Sigma}_q,\tilde{\rho}]|e\rangle \;=\; -\frac{1}{2}\sum_{e'}\tilde{\rho}_{ge'}\sum_{g'}\sum_q f_{g'e,q}\,f_{g'e',q}$$

If we write $f_{ge,q}$ more concisely as $f_{ge,q}=(-1)^{F_e+J_g+1+I}\sqrt{S_{F_gF_e}}\,\langle F_g,M_g|F_e,M_e;1,q\rangle$ with

$$S_{F_gF_e}=(2\,F_e+1)\,(2\,J_g+1)\left\{\begin{matrix} J_g & J_e & 1\\ F_e & F_g & I\end{matrix}\right\}^2$$

the sum $\sum_{g'}\sum_q f_{g'e,q}\, f_{g'e',q}$ can be factorized as

$$\sum_{F'_g} (-1)^{F_e+J_g+1+I+F'_e+J_g+1+I}\sqrt{S_{F'_g F_e}\, S_{F'_g F'_e}}\sum_{M'_g}\sum_q \langle F'_g, M'_g | F_e, M_e; 1, q\rangle\, \langle F'_g, M'_g | F'_e, M'_e; 1, q\rangle$$

$$= \sum_{F'_g}\cdots\sum_{M'_g}\sum_q (-1)^{F_e-1+M'_g}\sqrt{2\,F'_g+1}\begin{pmatrix} F_e & 1 & F'_g \\ M_e & q & -M'_g\end{pmatrix}(-1)^{F'_e-1+M'_g}\sqrt{2\,F'_g+1}\begin{pmatrix} F'_e & 1 & F'_g \\ M'_e & q & -M'_g\end{pmatrix}$$

(CB coeffs $\to$ 3j symbols so that it's easier to intervert columns)

$$= \sum_{F'_g}\cdots\cdots (-1)^{F_e+F'_e}(2\,F'_g+1)\sum_{M'_g}\sum_q (-1)^{2M'_g}\begin{pmatrix} F_e & 1 & F'_g \\ M_e & q & -M'_g\end{pmatrix}\begin{pmatrix} F'_e & 1 & F'_g \\ M'_e & q & -M'_g\end{pmatrix}$$

(even column permutation)

$$= \sum_{F'_g}\cdots\cdots (-1)^{F_e+F'_e}(2\,F'_g+1)\,(-1)^{2F'_g}\sum_{M''_g}\sum_q \begin{pmatrix} 1 & F'_g & F_e \\ q & M''_g & M_e\end{pmatrix}\begin{pmatrix} 1 & F'_g & F'_e \\ q & M''_g & M'_e\end{pmatrix}$$

($2\,M'_g$ has the same parity than $2\,F'_g$; replacing $-M'_g$ by M''_g)

$$= \sum_{F'_g}\cdots\cdots (-1)^{F_e+F'_e}(2\,F'_g+1)\,(-1)^{2F'_g}\,\frac{\delta_{F'_e F_e}\,\delta_{M'_e M_e}\,\{\,1\ \ F'_g\ \ F_e\,\}}{2\,F_e+1}$$

(orthogonality relation of 3j symbols on the initial basis sector; $\{\,\cdot\ \ \cdot\ \ \cdot\,\}$ is the triangular condition)

$$= \sum_{F'_g} (-1)^{2F_e+2J_g+2I+2F_e+2F'_g}\, S_{F'_g F_e}\,\frac{2\,F'_g+1}{2\,F_e+1}\,\delta_{F'_e F_e}\,\delta_{M'_e M_e}$$

(the triangular condition $\{\,1\ \ F'_g\ \ F_e\,\}$ is already in the 6j symbol of $S_{F'_g F_e}$)

$$= \delta_{ee'}\sum_{F'_g} S_{F'_g F_e}\,\frac{2\,F'_g+1}{2\,F_e+1}$$

($4\,F_e$ is even; $2\,(J_g+I+2\,F'_g)$ has to be even too)

$$= \delta_{ee'}\sum_{F'_g}\frac{2\,F'_g+1}{2\,F_e+1}\,(2\,F_e+1)\,(2\,J_g+1)\begin{Bmatrix} J_g & J_e & 1 \\ F_e & F_g' & I\end{Bmatrix}^2$$

$$= \delta_{ee'}\,(2\,J_g+1)\sum_{F'_g}(2\,F'_g+1)\begin{Bmatrix} J_g & I & F_g' \\ F_e & 1 & J_e\end{Bmatrix}^2$$

(column and up-down swapping by symmetry of 6j symbols)

$$= \delta_{ee'}\,(2\,J_g+1)\,\frac{\delta_{J_e J_e}\,\{\,J_g\ \ 1\ \ J_e\,\}\,\{\,F_e\ \ I\ \ J_e\,\}}{2\,J_e+1}$$

(orthogonality relation of 6j symbols)

$$= \delta_{ee'}\,\frac{2\,J_g+1}{2\,J_e+1}$$

(the triangular condition is satisfied by definition / optical selection rules on J's and F's)

Maybe this result is actually well known and expected from the addition of three angular momenta. Finally, we obtain that

$$\hbar\,\Gamma\,\tfrac{2\,J_e+1}{2\,J_g+1}\sum_q \langle g|\mathcal{D}[\mathbf{\Sigma}_q,\tilde\rho]|e\rangle = -\tfrac{1}{2}\,\hbar\,\Gamma\,\tfrac{2\,J_e+1}{2\,J_g+1}\sum_{e'}\tilde\rho_{ge'}\,\delta_{ee'}\,\tfrac{2\,J_g+1}{2\,J_e+1}$$

$$= -\tfrac{1}{2}\,\hbar\,\Gamma\,\tilde\rho_{ge}$$

Gathering all three terms, we obtain (3).

Coherences ?

Note that if we would keep coherences in $\langle g|[\tilde{\boldsymbol{H}}_{\text{atom-light}},\tilde{\rho}]|e\rangle$, the adiabatic approximation would yield

$$\tilde{\rho}_{ge}\approx-\tfrac{\mathrm{i}}{2}\frac{1}{\mathrm{i}\,\Delta_{eg}+\Gamma/2}\sum_{q'}\Omega_{q'}^{*}\left(\sum_{e'}f_{ge',q'}\,\tilde{\rho}_{e'e}-\sum_{g'}f_{g'e,q'}\,\tilde{\rho}_{gg'}\right)$$

Inserting that into $\partial_t\,\tilde{\rho}_{gg}$, we would end up with terms like $\sum_q\sum_{q'}\Omega_q\,\Omega_{q'}^{*}\,f_{ge,q}\,(f_{ge',q'}\,\tilde{\rho}_{e'e}-f_{g'e,q'}\,\tilde{\rho}_{gg'})$. With a single field mode ($\Omega_q=\Omega_{q_0}\,\delta_{qq_0}$), this simplifies to $\Omega_{q_0}\,\Omega_{q_0}^{*}\,f_{ge,q_0}\,(f_{ge',q_0}\,\tilde{\rho}_{e'e}-f_{g'e,q_0}\,\tilde{\rho}_{gg'})$, and again this removes M_F coherences, leaving only F coherences that we can most often approximate out.

But if it is not the case, the whole system becomes a huge coupled mess of equations because we have to keep track of the coherences, and eventually compute them too in the adiabatic approximation.

3.3. $\partial_t\,\tilde{\rho}_{ee}$

The derivation of $\tilde{\rho}_{ee}$ is very similar to the one of $\tilde{\rho}_{gg}$; only the decay term derivation is different. Similar to (9), we obtain

$$\langle e|\mathcal{D}[\boldsymbol{\Sigma}_q,\tilde{\rho}]|e\rangle\;=\;-\tfrac{1}{2}\sum_{g',e'}f_{g'e',q}\,f_{g'e,q}\,(\tilde{\rho}_{e'e}+\tilde{\rho}_{ee'})$$

Dropping e-e' coherences, we get that

$$\begin{aligned}\sum_q\Gamma\,\tfrac{2\,J_e+1}{2\,J_g+1}\,\langle e|\mathcal{D}[\boldsymbol{\Sigma}_q,\tilde{\rho}]|e\rangle\;&=\;\sum_q\Gamma\,\tfrac{2\,J_e+1}{2\,J_g+1}\left(-\tfrac{1}{2}\right)\sum_{g'}f_{g'e,q}^{2}\,2\,\tilde{\rho}_{ee}\\&=\;-\Gamma\,\tilde{\rho}_{ee}\sum_q\sum_g\tfrac{2\,J_e+1}{2\,J_g+1}\,f_{ge,q}^{2}\\&=\;-\Gamma\,\tilde{\rho}_{ee}\sum_q\sum_g\mathcal{F}_{eg,q}\end{aligned}$$

The last sum of branching ratios $\mathcal{F}_{eg}$ over F_g,M_g simplifies to 1. Indeed, very similarly to the treatment of the decay term of $\partial_t\,\tilde{\rho}_{ge}$ above,

$$\begin{aligned}\sum_q\sum_g\mathcal{F}_{eg,q}\;&=\;\sum_q\sum_{F_g,M_g}(2\,F_e+1)\,(2\,J_e+1)\begin{Bmatrix}J_g&J_e&1\\F_e&F_g&I\end{Bmatrix}^2(2\,F_g+1)\begin{pmatrix}F_e&1&F_g\\M_e&q&-M_g\end{pmatrix}^2\\\text{[}e\text{-}g\text{ exchange]}\;&=\;(2\,J_e+1)\sum_{F_g}(2\,F_e+1)\begin{Bmatrix}J_g&J_e&1\\F_e&F_g&I\end{Bmatrix}^2(2\,F_g+1)\sum_{M_g,q}\begin{pmatrix}1&F_g&F_e\\q&-M_g&M_e\end{pmatrix}^2\\\text{[orthogonality]}\;&=\;(2\,J_e+1)\sum_{F_g}(2\,F_e+1)\begin{Bmatrix}J_g&J_e&1\\F_e&F_g&I\end{Bmatrix}^2(2\,F_g+1)\,\frac{\delta_{F_eF_e}\,\delta_{M_eM_e}\,\{\;1\;\;F_g\;\;F_e\;\}}{2\,F_e+1}\\\begin{matrix}\text{[triang. cond.}\\\text{already satisfied]}\end{matrix}\;&=\;(2\,J_e+1)\sum_{F_g}(2\,F_g+1)\begin{Bmatrix}J_g&J_e&1\\F_e&F_g&I\end{Bmatrix}^2\\\text{[6j orthogonality]}\;&=\;(2\,J_e+1)\,\frac{\delta_{J_eJ_e}\,\{\;J_g\;\;1\;\;J_e\;\}\,\{\;F_e\;\;I\;\;J_e\;\}}{2\,J_e+1}\\&=\;1\quad\text{[triangular condition is satisfied by definition / optical selection rules on }J\text{'s and }F\text{'s]}\end{aligned}\tag{13}$$

Bibliography

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