

PyLCP: A package for computing complicated laser cooling physics

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Abstract

A submitted program is expected to be of benefit to other physicists or physical chemists, or be an exemplar of good programming practice, or illustrate new or novel programming techniques which are of importance to some branch of computational physics or physical chemistry.

Acceptable program descriptions can take different forms. The following Long Write-Up structure is a suggested structure but it is not obligatory. Actual structure will depend on the length of the program, the extent to which the algorithms or software have already been described in literature, and the detail provided in the user manual.

Your manuscript and figure sources should be submitted through the Elsevier Editorial System (EES) by using the online submission tool at <http://www.ees.elsevier.com/cpc>.

In addition to the manuscript you must supply: the program source code; job control scripts, where applicable; a README file giving the names and a brief description of all the files that make up the package and clear instructions on the installation and execution of the program; sample input and output data for at least one comprehensive test run; and, where appropriate, a user manual. These should be sent, via email as a compressed archive file, to the CPC Program Librarian at cpc@qub.ac.uk.

Keywords: keyword1; keyword2; keyword3; etc.

PROGRAM SUMMARY

Program Title: PyLCP

Licensing provisions(please choose one): GPLv3

Programming language: Python

Nature of problem(approx. 50-250 words):

Solution method(approx. 50-250 words):

Additional comments including Restrictions and Unusual features (approx. 50-250 words):

[1] Reference 1

[2] Reference 2

[3] Reference 3

* Items marked with an asterisk are only required for new versions of programs previously published in the CPC Program Library.

1. Introduction

The calculation of various quantities in laser cooling is hard.

Here we introduce a python-based program that computes the movement of atoms or molecules with complex level structures in arbitrary laser fields, magnetic fields. The program is broken down

2. The optical Bloch equations

Here, we consider the generic problem of coupling many states together using laser light. We group these states into manifolds. A manifold of states is a collection of states that are degenerate or nearly degenerate, e.g., the 2S_0 states of an alkali atom, the ro-vibrational states of molecules, etc. We denote the n th state in the i th manifold by $|i, n\rangle$.

The full Hamiltonian is given by

$$\hat{H} = \hat{H}_{\text{atom}} + \hat{H}_{\text{field}} - \hat{\mathbf{d}} \cdot \hat{\mathbf{E}} - \hat{\boldsymbol{\mu}} \cdot \hat{\mathbf{B}}. \quad (1)$$

The field component of the Hamiltonian is given by

$$\hat{H}_{\text{field}} = \int \left(\epsilon_0 \hat{\mathbf{E}} + \frac{\hat{\mathbf{B}}}{2} \right) dV \quad (2)$$

where $\hat{\mathbf{E}}$ is the electric field operator, $\hat{\mathbf{B}}$ is the magnetic field operator. The atomic Hamiltonian,

$$\hat{H}_{\text{atom}} = \frac{P^2}{2M} + \hat{H}_{\text{internal}} \quad (3)$$

where $\hat{H}_{\text{internal}}$ describes the internal structure of the atom. In general, it has the form

$$\hat{H}_{\text{internal}} = \sum_{i,n} [\hbar(\omega_{m,i} + \omega_n) |i, n\rangle \langle i, n|] \quad (4)$$

where $\omega_{m,i}$ is the offset frequency of the i th manifold and ω_n is the n th's state frequency relative to $\omega_{m,i}$. Manifolds are connected only through $\mathbf{d} \cdot \mathbf{E}$ component of the Hamiltonian; $\hat{H}_{m,i}$ and $\hat{\boldsymbol{\mu}}$ only act on the subspace of each manifold.

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Our goal is to find the evolution of the atom operators $\rho_{nm}^{ij} = |i, n\rangle \langle j, m|$. In the Heisenberg picture, the operators \hat{O} evolve as

$$\frac{\partial \hat{O}}{\partial t} = \frac{i}{\hbar} [\hat{H}, \hat{O}]. \quad (5)$$

If the fields were treated classically, this equation would have to be amended in order to take into account decays. Instead, if we quantize the electric fields, derive the equations of motion, and then apply appropriate radiation reaction approximations, we can derive the full optical Bloch equations with decay from Eq. 5. The magnetic field \mathbf{B} is assumed to be a classical field; we will not consider quantizing it. We must pay special focus to the electric field, however, for it both shifts the internal Hamiltonian when transformed into the necessary rotating frame(s) and creates the necessary decay channels.

For the electric field, \mathbf{E} could be comprised of multiple modes. We group those modes by manifold indices $i \rightarrow j$. Within each manifold, in addition to the mean frequency $\omega_{i \rightarrow j}$, there could then be multiple modes, which we then index by ω_k . Thus,

$$\mathbf{E} = \sum_{i,j,k} \mathbf{E}_{i \rightarrow j,k} e^{-i(\omega_{i \rightarrow j} + \omega_k)t} + \mathbf{E}_{i \rightarrow j,k}^\dagger e^{i(\omega_{i \rightarrow j} + \omega_k)t} \quad (6)$$

Here \mathbf{E} represents a destruction operator of the mode $i \rightarrow j, k$. We similarly expand the dipole operator

$$\mathbf{d} = \sum_{i,j,n,m} \mathbf{d}_{nm}^{ij} |i, n\rangle \langle j, m| + |j, m\rangle \langle i, n| (\mathbf{d}_{nm}^{ij})^\dagger. \quad (7)$$

In general, the d_{nm}^{ij} are dependent on reduced matrix elements and Clebsch-Gordon coefficients that determine the transitions between manifolds i and j . We will not consider any specific form of d_{nm}^{ij} , but instead focus on deriving the optical Bloch equations for any generic form.

For each manifold, we will assume that all states in that manifold rotate at a preferred frequency $|i, n\rangle \rightarrow e^{i\omega_{ri}t} |i, n\rangle$. We choose the ω_{ri} such that their differences $\omega_{r,j} - \omega_{r,i} \approx \omega_{m,j} - \omega_{m,i} \approx \omega_{i \rightarrow j}$ for all combinations of i and j . This choice places each manifold into an appropriate rotating frame. In particular, we define $\delta_{i,n}^H = \omega_{r,i} - (\omega_{m,i} + \omega_n)$. Under this transformation of the state vectors, the internal Hamiltonian becomes

$$\hat{H}_{\text{internal}} = \sum_{i,n} -\hbar \delta_{i,n}^H |i, n\rangle \langle i, n|, \quad (8)$$

incorporating the shift into the rotating frame into the internal Hamiltonian. We also define $\delta_{i \rightarrow j,k}^L = (\omega_{i \rightarrow j} + \omega_k) - (\omega_{r,j} - \omega_{r,i})$. Making the rotating wave approximation (neglecting terms oscillating at optical frequencies), we find

$$\mathbf{d} \cdot \mathbf{E} = \sum_{i,j,k,n,m} [(\mathbf{d}_{nm}^{ij} \cdot \mathbf{E}_{i \rightarrow j,k}^\dagger) e^{-i\delta_{i \rightarrow j,k}^L t} |i, n\rangle \langle j, m| + |j, m\rangle \langle i, n| (\mathbf{d}_{nm}^{ij})^\dagger \cdot \mathbf{E}_{i \rightarrow j,k} e^{i\delta_{i \rightarrow j,k}^L t}]. \quad (9)$$

I really dislike ending a paragraph with an equation.

It is perhaps instructive to consider a couple examples of this construction of the rotating frame(s). Consider first a standard

two level system, with indices $i = g$ and $j = e$ and energies $\omega_g = 0$ and ω_e , being driven by a single electric field with total frequency ω . (Here, we drop the k, n , and m subscripts.) Let us take $\omega_{r,g} = 0$ and $\omega_{r,e} = \omega_r$. Then the detuning on the Hamiltonian, $\delta_e^H = \omega_r - \omega_e$ and $\delta_{g \rightarrow e}^L = \omega - \omega_r$. The total detuning of the laser from the excited state is then given by $\delta = \delta_{g \rightarrow e}^L + \delta_e^H = \omega - \omega_e$. In this way, we can split the detuning between lasers and Hamiltonian in whichever way yields best computational efficiency for the particular problem at hand.

Next, consider a three-level manifold Λ -system with a single state in each manifold. Let us label the manifolds as g, r and e in order of overall energy, and we drop the unnecessary substate subscripts. We address this system with two lasers, one tuned closely to $g \rightarrow e$ with frequency $\omega_{g \rightarrow e}$ and the other tuned closely to $r \rightarrow e$ with frequency $\omega_{r \rightarrow e}$. We now choose $\omega_{r,g} = 0$, and the relevant detunings are then

$$\delta_g^H = 0 \quad (10)$$

$$\delta_e^H = \omega_{r,e} - \omega_e \quad (11)$$

$$\delta_r^H = \omega_{r,r} - \omega_r \quad (12)$$

$$\delta_{g \rightarrow e}^L = \omega_{g \rightarrow e} - \omega_{r,e} \quad (13)$$

$$\delta_{r \rightarrow e}^L = \omega_{r \rightarrow e} - (\omega_{r,e} - \omega_{r,r}) \quad (14)$$

By choosing $\omega_{r,e} = \omega_{g \rightarrow e}$ and $\omega_{r,r} = \omega_{g \rightarrow e} - \omega_{r \rightarrow e}$, one recovers the textbook example of the three level system with detunings shifted onto the Hamiltonian. As with the two level system above, one can split the detunings between lasers and Hamiltonian in whichever way yields best computational efficiency for the particular problem at hand.

Before applying the radiation reaction approximation, we must first find the equations of motion. Inserting Eq. 9 into Eq. 5, and using $\rho_{nm}^{ij} \rho_{pq}^{kl} = \rho_{nq}^{il} \delta_{jk} \delta_{mp}$, where δ_{ij} is the Kronecker delta function

$$\frac{\partial \rho_{nm}^{ij}}{\partial t} = \sum_{jklmnpq} +i\sigma_{kl}^{ab} (\mathbf{d}_{ij}^{12} \cdot \mathbf{E}^\dagger) \sigma_{ij}^{12} + i\sigma_{kl}^{ab} \sigma_{ji}^{21} ([\mathbf{d}_{ij}^{12}]^\dagger \cdot \mathbf{E})$$

$$\sigma_{kl}^{ab} = i[H, \sigma_{kl}^{ab}] = -i(\mathbf{d}_{ij}^{12} \cdot \mathbf{E}^\dagger) \sigma_{ij}^{12} \sigma_{kl}^{ab} - \sigma_{ji}^{21} ([\mathbf{d}_{ij}^{12}]^\dagger \cdot \mathbf{E}) \sigma_{kl}^{ab} + i\sigma_{kl}^{ab} (\mathbf{d}_{ij}^{12} \cdot \mathbf{E}^\dagger) \sigma_{ij}^{12} + i\sigma_{kl}^{ab} \sigma_{ji}^{21} ([\mathbf{d}_{ij}^{12}]^\dagger \cdot \mathbf{E})$$

To determine the decays, we must now apply a radiation reaction approximation. Classically, the radiation reaction field is

$$\mathbf{E}_{RR} = \frac{1}{6\pi\epsilon_0 c^3} \frac{d^3 \mathbf{d}}{dt^3}. \quad (17)$$

Classically, the frequency of oscillation of the reaction field will be dominated by the frequency closest to resonance. We make the approximation that the For each mode ω_k in the manifold,

$$\begin{aligned} \frac{\partial \rho_{ij}}{\partial t} &= [-i(\mathbf{d}_{ki} \cdot \mathbf{E}^\dagger) \rho_{kj} - i\rho_{kj} (\mathbf{d}_{ki}^\dagger \cdot \mathbf{E}) + i(\mathbf{d}_{jk} \cdot \mathbf{E}^\dagger) \rho_{ik} + \rho_{ik} (\mathbf{d}_{jk}^\dagger \cdot \mathbf{E}) \\ &= [-\mathbf{d}_{ki} \cdot \mathbf{d}_{lm}^\dagger \rho_{lm} \rho_{kj} + \rho_{kj} \mathbf{d}_{ki}^\dagger \cdot \mathbf{d}_{lm} \rho_{lm} + \mathbf{d}_{jk} \cdot \mathbf{d}_{lm}^\dagger \rho_{lm} \rho_{ik} - \rho_{ik} \mathbf{d}_{jk}^\dagger \cdot \mathbf{d}_{lm} \rho_{lm} \\ &= [-\mathbf{d}_{ki} \cdot \mathbf{d}_{lk}^\dagger \rho_{lj} + \mathbf{d}_{ki}^\dagger \cdot \mathbf{d}_{jl} \rho_{kl} + \mathbf{d}_{jk} \cdot \mathbf{d}_{li}^\dagger \rho_{lk} - \mathbf{d}_{jk}^\dagger \cdot \mathbf{d}_{il} \rho_{il}]] \end{aligned}$$

2.1. Representation of the Hamiltonian

In `pylcp`, we represent this Hamiltonian as a series of blocks, with each block containing a manifold of states (e.g., Zeeman sub-levels or a manifold of hyperfine states). A completely generic basis set vector can then be written as

$$|\phi\rangle = \begin{pmatrix} |l_1\rangle \\ \vdots \\ |l_{N_l}\rangle \\ |n_1\rangle \\ \vdots \\ |n_{N_n}\rangle \end{pmatrix}, \quad (21)$$

where $|l_i\rangle$ are the eigenstates of the first (ground) manifold and $|n_k\rangle$ are the eigenstates of the most excited manifold. With this basis vector, the term $\boldsymbol{\mu}_l \cdot \mathbf{B}$ is the field dependent term that mixes states within a given manifold l and $\vec{d}_{lm} \cdot \mathbf{E}_{lm}$ is the field dependent term that couples states of different manifolds. Using labels $i = g, e$ for the two extreme manifolds, the Hamiltonian blocks look like

$$H_{\text{atom}} = \begin{pmatrix} (H_g - \boldsymbol{\mu}_g \cdot \mathbf{B}) & \cdots & (\mathbf{d}_{ge} \cdot \mathbf{E}_{ge}) \\ \vdots & \ddots & \vdots \\ (\mathbf{d}_{ge}^* \cdot \mathbf{E}_{ge}^*) & \cdots & (H_e + \boldsymbol{\mu}_e \cdot \mathbf{B}) \end{pmatrix}, \quad (22)$$

where each element in the matrix H_g, H_e , etc. is itself a matrix. In general, the the electric fields driving transitions between manifolds are distinct, which is why the electric field gains a specific label in Eq. 22.

To specify the problem, the user defines a Hamiltonian like Eq. (5) by providing the requisite H_0 , $\boldsymbol{\mu}_l$, and \mathbf{d}_{lm} and combining them in the `hamiltonian` class. The class creates and stores the block structure of the Hamiltonian, and also contains methods that are useful for its manipulation. The vectors $\boldsymbol{\mu}$ and \mathbf{d} are represented in spherical polar coordinates, allowing for translation into σ^\pm circular polarization and π polarizations. For these vectors, we denote the component q as d^q .

In its construction, `pylcp` assumes a that the Hamiltonian is written in a co-rotating frame with a frequency close to that of the laser light, with the number of rotating frames equal to $N_m - 1$. The electric field has the form

$$\mathbf{E}_{lm} = \frac{1}{2} \hat{\mathbf{e}}(r, t) E_{lm}(r, t) e^{i\mathbf{k}(r, t) \cdot \mathbf{r} - i\Delta t + \phi(t)} \quad (23)$$

where the complex conjugate term is neglected by the rotating wave approximation. The user specifies all components of the laser field: the polarization vector $\hat{\mathbf{e}}(r, t)$, the amplitude $E_{lm}(r, t)$, the \mathbf{k} vector, the average detuning between the rotating frame and optical frequency Δ , and any potential phase modulation of the laser beam $\phi(t)$ in the `LaserBeam` class.

Finally, the magnetic field \mathbf{B} is specified simply as a user-defined function of space and time. With all of these components, the optical Bloch equations and/or rate equations can be constructed for a given set of parameters.

2.2. The optical Bloch equations

The optical Bloch equations are equations that govern the evolution of the density matrix with the requisite decays. They are equations that consider the evolution of a given density operator $\rho_{l,m_j} = |l_i\rangle\langle m_j|$. The master equation for their evolution, in the Heisenberg picture, is

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} [\rho, H] \quad (24)$$

Specifically, for the $|i_n\rangle\langle j_m|$ component,

$$\dot{\rho}_{nm}^{ij} = -\frac{i}{\hbar} [\rho, H]_{i_n, j_m} = -\frac{i}{\hbar} [\rho_{np}^{ik} H_{pm}^{kj} - \rho_{pm}^{ki} H_{np}^{ik}] \quad (25)$$

where Einstein summation is assumed. We include decay through the radiation-reaction approximation. In this approximation, we take the dipole operator

$$\mathbf{E}_{ij} = \mathbf{E}_{0,ij} + i \sum_{nm} \frac{k_{ij}^3}{6\pi\epsilon_0} \mathbf{d}_{nm}^{ij} \rho_{nm}^{ij}. \quad (26)$$

Then, for the d_{nm}^{ij} , the operator becomes

$$\dot{\rho}_{nm}^{ij} = -\frac{i}{\hbar} [\rho_{np}^{ij} d_{pm}^{ij} (d_{mr}^{ij})^\dagger \rho_{mr}^{ji} - \rho_{pm}^{ki} H_{np}^{ik}] \quad (27)$$

The relevant form the Linblad super operator becomes

$$\rho \mathcal{L} \rho = i \frac{k^3}{3\pi\epsilon_0} \boldsymbol{\mu}_{nm}^{ij} \rho_{nm}^{ij} \quad (28)$$

The package `pylcp` constructs the first term from an arbitrary Hamiltonian (Eq. 22) by directly computing the commutator. Programmatically, it represents the full density matrix as a vector represents the complicated temporal evolution as a matrix, with components that depend on \mathbf{E}_{lm} and \mathbf{B} .

The particle's semiclassical motion can be calculated through

$$\ddot{\mathbf{r}} = -\frac{1}{m} \{\nabla H\} + \mathbf{a} = \frac{1}{m} \{\nabla(\mathbf{d} \cdot \mathbf{E} + \boldsymbol{\mu} \cdot \mathbf{B})\} + \mathbf{a} \quad (29)$$

where $\ddot{\mathbf{r}}$ is the acceleration of the particle, \mathbf{a} is a constant acceleration, and [...].

For `pylcp`, $\hbar = 1$ so that angular frequencies and energies are equivalent. In practice, the user specifies H_0/\hbar , rather than H_0 . When forming a complete Hamiltonian using the `hamiltonian` class, the user has the ability to set the units by specifying base units of length x_0 , time t_0 , magnetic field B_0 and mass m . For $\mathbf{d} \cdot \mathbf{E}$, there are two properties to quantify: the associated wavevector $k_{ij}x_0$, important for random forces, and the decay rate Γt_0 . For the magnetic Hamiltonian, the user has to specify the Bohr magneton in their preferred units, $\mu_B B_0/\hbar t_0$, in order to determine the preferred units of magnetic field. By default, a two-manifold system uses units $t_0 = 1/\Gamma$ and $x_0 = 1/k$.

For lasers, we specify everything in terms of the decay rate of the excited state Γ and saturation intensity I_{sat}

$$I_{\text{sat}} = \frac{\pi \hbar c \Gamma}{3\lambda^3} = \frac{2\pi^2 \hbar \omega^3 c \Gamma}{3(2\pi)^3 c^3} = \frac{\hbar \omega^3 \Gamma}{12\pi c^2} \quad (30)$$

the result being that the electric field $E_{lm}(r, t) = \sqrt{I/2I_{\text{sat}}} = \sqrt{\beta/2}$. Included Hamiltonians normalize each element in the $(d \cdot E)_{ij}$ Hamiltonian to Γ_{ij} so that a laser with $I/I_{\text{sat}} = 1$ driving the transition yields a Rabi rate of $\Gamma_{ij}/\sqrt{2}$. Likewise, included Hamiltonians also return μ without the spin, i.e., $\mu = \mu_B \mathbf{F}$, where \mathbf{F} is some generalized spin operator.

For the force, this leaves one additional unit to specify, the mass. If the user specifies the length, time, and magnetic field, then

$$\frac{x_0}{t_0^2} \ddot{\mathbf{r}}' = \frac{\hbar}{x_0 t_0 m} \{-\nabla' H'\} + \frac{x_0}{t_0^2} \mathbf{a}'. \quad (31)$$

The default unit selection for a two-manifold system is $t_0 = 1/\Gamma$ and $x_0 = 1/k$, which yields for the prefactor $\hbar/x_0 t_0 m = \hbar k \Gamma / m$. Further taking into account the units on the left hand side we define

$$\ddot{\mathbf{r}}' = \frac{\hbar t_0}{x_0 m} \{-\nabla' H'\} + \mathbf{a}' \quad (32)$$

which defines the ‘dimensionless’ mass as $x_0 m / \hbar t_0$.

The optical Bloch equations are symmetric under population exchange, i.e., $\rho_{l_i, m_j} = \rho_{m_j, l_i}^*$. The package `pylcp` takes advantage of this symmetry to transform the optical Bloch equations into , increasing speed.

2.3. The Rate Equations

In order to do a rate equation model, we first consider that we have a set of eigenstates and that the matrix \mathbf{H} is diagonal. We follow the construction of the rate equations from Ref. [?] and, to a lesser extent, the Ref. [?]. In the rate equation model, we consider three things. The first is the excitation rate caused by laser l

$$R_{ij}^l = \Gamma \left(\frac{\Omega_{ij}^l}{\Gamma} \right)^2 \frac{1}{1 + 4(\Delta - k \cdot v)^2 / \Gamma^2}, \quad (33)$$

and the decay rate out of the excited state Γ , and the branching ratio,

$$\Gamma_{e_j, g_i} = \Gamma \frac{|d_{ij}^{(q)}|^2}{\left(\sum_i |d_{ij}^{(q)}|^2 \right)}. \quad (34)$$

In general, we want to solve for $\dot{N}_{g/e, i}$. We will do this in units of Γ . For the ground states,

$$\dot{N}_i = - \sum_{jl} R_{ij}^l (N_i - N_j) + \sum_{jl} \Gamma_{ij} N_j \quad (35)$$

and for the excited states,

$$\dot{N}_j = -\Gamma N_j + \sum_{jl} R_{ij}^l (N_i - N_j) \quad (36)$$

We represent the rate equations as a matrix R such that

$$\dot{N} = RN \quad (37)$$

where $N = (N_{g_1}, \dots, N_{g_n}, N_{e_1}, \dots, N_{g_n})$.

2.4. Random Forces

In both the OBE and

2.5. Units

Units in `pylcp` are determined through the following master evolution equations for the OBEs:

$$\dot{\rho} = \frac{i}{\hbar} [\rho, H] \quad (38)$$

$$\ddot{\mathbf{r}} = -\frac{1}{m} \{\nabla H\} + \mathbf{a} = \frac{1}{m} \{\nabla(\mathbf{d} \cdot \mathbf{E} + \mu \cdot \mathbf{B})\} + \mathbf{a} \quad (39)$$

3. Additional details

3.1. A word about polarization

We have thusfar considered the polarization to be contained E_q^l . However, computing this quantity is not necessarily trivial. First, one takes the magnitude out realizing that is related to the intensity, i.e., $E_q^l = |E|^l \hat{e}_q^l$. Second, \hat{e}_q^l is defined in the rotational basis of the quantization axis, whereas for an actual laser beam, the polarization is defined via some other means.

In general, the polarization is a complex-three space vector, which therefore has 5 independent parameters. However, because light is a transverse wave and \hat{k} is defined (with two parameters), that reduces the number of parameters to three. We often think about the Poincare sphere or polarization ellipse at this stage which is defined by only two parameters, but there is a hidden parameter which is the orientation of the coordinate system in which the sphere or ellipse is defined.

For the purposes of computation of $\hat{e}_q \cdot \hat{e}_l$, there seems to be multiple ways to define the coordinate system. Consider, for example, defining the polarization in terms of right ϵ^+ and left ϵ^- polarized light. Then the most general polarization vector is $\epsilon_l = a\epsilon_l^+ + b\epsilon_l^-$, where a and b are complex constants and $|a|^2 + |b|^2 = 1$. There are thus three independent parameters that specify this polarization. Note that the phase between a and b determines the angle in the plane that a linear projection will point and that $\epsilon_l^+ = \sigma^+$ and $\epsilon_l^- = \sigma^-$ if the magnetic field is parallel to \hat{k} .

If \hat{k} is not parallel to the quantization axis, then we need to rotate one coordinate system (the laser and polarization) into the other (where the quantization is done). Imagine that the Euler angles doing the wrapping are α - β - γ in the z-y-z reference. In this case, the vector is rotated through a Wigner rotation matrix, whose elements are given by

$$D_{m'm}^j(\alpha, \beta, \gamma) = \langle jm' | \mathcal{R}(\alpha, \beta, \gamma) | jm \rangle = e^{im'\alpha} d_{m'm}^j(\beta) e^{-im\gamma} \quad (40)$$

where $d_{m'm}^j$ is the small Wigner rotation matrix. At first glance, the diagonal nature of matrix for α and γ seems to imply that those angle do not play an important role; all of the relevant physics is simply related to β . Alternatively, this would imply that the phase angle between the σ^+ and σ^- components of the unrotated light basis. We can explicitly construct the matrix

element, using $d_{m'm}^j$ from Wikipedia and the relationship $d_{m'm}^j = (-1)^{m-m'} d_{mm'}^j = d_{m,m'}^j$.

$$\mathcal{D} = \begin{pmatrix} \frac{1+\cos\beta}{2} e^{-i\alpha+i\gamma} & \frac{\sin\beta}{\sqrt{2}} e^{-i\alpha} & \frac{1-\cos\beta}{2} e^{-i\alpha-i\gamma} \\ -\frac{\sin\beta}{\sqrt{2}} e^{i\gamma} & \cos\beta & -\frac{\sin\beta}{\sqrt{2}} e^{-i\gamma} \\ \frac{1-\cos\beta}{2} e^{i\alpha+i\gamma} & \frac{\sin\beta}{\sqrt{2}} & \frac{1+\cos\beta}{2} e^{i\alpha-i\gamma} \end{pmatrix} \quad (41)$$

This clearly tells a more complicated story. The simplest observation might be that one of the Euler angles does not matter, at least for the rate equation model. In that case, we will square each component and the common phase $e^{\pm i\alpha}$ will drop out. This makes sense, as the rotation into the quantization axis should only have two angles, the third being degenerate because it is an axis not a full 3-D coordinate system.

Consider the final π ($m' = 0$) component. An input beam with equal amounts of σ^+ and σ^- defines linear polarization, and the relative phase between the two components defines where that points relative to some other axis, defined by the overall phase. If this differential phase is χ , then the π component would be given by

$$\hat{\epsilon}_l \cdot \hat{\epsilon}_0 = -\frac{\sin\beta}{\sqrt{2}} \cos(\gamma + \chi). \quad (42)$$

This dependence on the phase angle makes intuitive sense. Consider a linearly-polarized wave moving in $+\hat{x}$ and magnetic field along \hat{z} . If the phase angle was such that the two circular components added to produce linear polarization along \hat{z} , the polarization would be completely π . On the other hand, if the phase angle was such that the linear polarization was along \hat{y} , then in the spherical basis we would have equal amounts of σ^+ and σ^- light (with a phase angle in between).

None of this gets us closer to the final result, but it is interesting.

4. Examples

Optical pumping.