Polarization phase-contrast imaging

In this document we will introduce the technique of polarization phase-contrast imaging (PPCI), which is used at various stages of our experiment to measure the in-situ column density of the dilute gas. The general goal of atom cloud imaging is to obtain a quantitative measure of the density distribution of the sample by looking at the intensity, phase and polarization profiles of light that has gone through it. The widely used absorption imaging technique looks at the intensity attenuation profile for a resonant, or nearly resonant laser beam. On the other hand, light with a larger detuning is used for PPCI; rather than looking for attenuation of the intensity, the information about the density of the gas can be extracted from the phase profile of the transmitted beam. The treatment is separated into four parts, first we calculate how the light is affected by the polarized atomic cloud. This is done classicaly, using Maxwell's equations and assuming that we know the polarization of the medium. In the second part we calculate the general polarization of the atomic cloud, which results from the perturbation on the atoms by the oscillating electric field of the light. This culminates in the determination of the electric susceptibility tensor. In the third part we evaluate the elements of the suceptibility tensor for the atomic structure that is of interest in our application, namely a spin-mixture of ^{6}Li atoms at high magnetic field. We then proceed to derive expressions that relate the phase profile of the transmitted beam to the column density of the cloud. In the fourth part we examine the experimental setup which allows one to convert the phase profile into an intensity profile that can be recorded on a CCD camera.

1 Transmission of light through a polarized gas

The electric field of the light that traverses the atomic cloud can be written in general form (see Sec. 6.1 of [1]) as

$$\boldsymbol{\mathcal{E}}(\boldsymbol{r},t) = \mathrm{Re}\{\mathcal{E}_0 e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega t+\phi)}[(\cos\alpha\cos\epsilon - i\sin\alpha\sin\epsilon)\hat{\boldsymbol{e}}_1 + (\sin\alpha\cos\epsilon + i\cos\alpha\sin\epsilon)\hat{\boldsymbol{e}}_2]\} \tag{1}$$

where $\hat{\boldsymbol{e}}_{1,2}$ are two orthogonal unit vectors perpendicular to the wave vector $\boldsymbol{k}$ and perpendicular to each other, $\mathcal{E}_0$ is the electric field amplitude, ϕ is an overall phase, α is the angle between the major axis of the polarization ellipse and the $\hat{\boldsymbol{e}}_1$ axis, and ϵ is the arctangent of the ratio of the minor to the major axis of the polarization ellipse.

The wave equation in a dielectric medium can be obtained from Maxwell's equations (see Sec. 10.1 of [1]), and is given by

$$\nabla^2\boldsymbol{\mathcal{E}} - \frac{1}{c^2}\frac{\partial^2\boldsymbol{\mathcal{E}}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2\mathbf{P}}{\partial t^2} \tag{2}$$

here the medium polarization $\mathbf{P}$ (which is induced by the light) corresponds to the dipole moment per unit volume. If we know the dipole moment of a single atom $\langle d\rangle$ then

$$\mathbf{P} = n\langle d\rangle \tag{3}$$

where n is the density of the gas.

Just as the electric field was parameterized in Eq. 1, the same can be done with the polarization in terms of the four real parameters P_i as

$$\mathbf{P} = \mathrm{Re}\{e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega t+\phi)}[(P_1 - iP_2)\hat{\boldsymbol{e}}_1 + (P_3 - iP_4)\hat{\boldsymbol{e}}_2]\} \tag{4}$$

Using the oscillatory time dependence of $\boldsymbol{\mathcal{E}}$ and $\mathbf{P}$ the wave equation is reduced to

$$\frac{\partial^2\boldsymbol{\mathcal{E}}}{\partial\ell^2} + k^2\boldsymbol{\mathcal{E}} = -4\pi k^2\mathbf{P} \tag{5}$$

where ℓ is the distance along the light propagation direction.

The parameterized electric field and polarization are plugged into the wave equation, and one proceeds to neglect terms that are of second order in the derivatives of the electric field parameters. This justifiable as long as the fractional change of the parameters is small over one wavelength of the light, which is certainly the case for our dilute atom cloud. The resulting expressions for the change of the field parameters per unit distance are:

$$\frac{1}{\mathcal{E}_0}\frac{d\mathcal{E}_0}{d\ell} = \frac{2\pi\omega}{\mathcal{E}_0 c}[-P_1\sin\alpha\sin\epsilon + P_4\sin\alpha\cos\epsilon + P_2\cos\alpha\cos\epsilon + P_3\cos\alpha\sin\epsilon] \tag{6}$$

$$\frac{d\varphi}{d\ell} = \frac{2\pi\omega}{\mathcal{E}_0 c}\sec 2\epsilon[P_1\cos\alpha\cos\epsilon + P_4\cos\alpha\sin\epsilon - P_2\sin\alpha\sin\epsilon + P_3\sin\alpha\cos\epsilon] \tag{7}$$

$$\frac{d\alpha}{d\ell} = \frac{2\pi\omega}{\mathcal{E}_0 c}\sec 2\epsilon[P_1\cos\alpha\sin\epsilon + P_4\cos\alpha\cos\epsilon - P_2\sin\alpha\cos\epsilon + P_3\sin\alpha\sin\epsilon] \tag{8}$$

$$\frac{d\epsilon}{d\ell} = -\frac{2\pi\omega}{\mathcal{E}_0 c}[P_1\sin\alpha\cos\epsilon + P_4\sin\alpha\sin\epsilon + P_2\cos\alpha\sin\epsilon - P_3\cos\alpha\cos\epsilon] \tag{9}$$

2 Polarization of the cloud

If we can calculate the electric dipole moment induced on one atom, $\langle \boldsymbol{d} \rangle$, and if we know the density of the cloud, $n(\vec{r})$, then we can simply write the induced polarization as

$$\mathbf{P}(\boldsymbol{r}) = \langle \boldsymbol{d} \rangle n(\boldsymbol{r}) \tag{10}$$

An ensemble of atoms (in which each atom may be in a different quantum state) is best described using the density matrix, which is defined as

$$\rho = \frac{1}{N}\sum_{i=1}^{N} |\psi_i\rangle\langle\psi_i| \tag{11}$$

where the i^{th} atom in the ensemble is in state $|\psi_i\rangle$. The expectation value of the electric dipole moment is then given by

$$\begin{aligned}\langle \boldsymbol{d} \rangle &= \mathrm{Tr}(\rho \boldsymbol{d}) \\ &= \sum_{mn} \rho_{mn}\langle n|\boldsymbol{d}|m\rangle\end{aligned} \tag{12}$$

Since the dipole operator only couples states of different parity, the terms that contribute to this sum correspond to m, n being a ground and excited state pair.

The equation of motion for the density matrix follows from the Schrodinger equation for the $|\psi_i\rangle$, it is known as the Liouville equation, which, including the spontaneous decay of the excited state, is [1]

$$i\hbar\dot{\rho} = [H,\rho] - \frac{i\hbar}{2}\{\hat{\Gamma},\rho\} + i\hbar\mathrm{Tr}(F\rho) \tag{13}$$

where $\hat{\Gamma}$ is the relaxation matrix, a diagonal matrix with the decay rate of each state on the diagonal (see Sec. 5.5 in [1]), and F is the spontaneous emission operator (see Sec. 12.1 in [1]). The hamiltonian H can be split into a diagonal part, H_0, which includes the level structure of the atom in the presence of a magnetic field, and the time dependent perturbation from the electric field $\hbar V$:

$$H = H_0 + \hbar V \tag{14}$$

The matrix element ρ_{ge}, where g, e represent a ground and an excited state respectively, satisfies

$$\begin{aligned}\dot{\rho}_{ge} &= \frac{1}{i\hbar}\langle g|[H_0,\rho]|e\rangle - i\langle g|[\hbar V,\rho]|e\rangle - \frac{1}{2}\langle g|\{\hat{\Gamma},\rho\}|e\rangle + \langle g|\mathrm{Tr}(F\rho)|e\rangle \\ &= -i\tilde{\omega}_{ge}\rho_{ge} - i\sum_p (V_{gp}\rho_{pe} - \rho_{gp}V_{pe}) + \sum_{rs} F^{sr}_{ge}\rho_{rs}\end{aligned} \tag{15}$$

where

$$\tilde{\omega}_{ge} = (E_g - E_e)/\hbar - i\Gamma_e/2 \tag{16}$$

We notice that $F^{sr}_{ge} = 0$, since F is only nonzero if the lower indices are both ground states and the upper indices are both excited states. So we are left with

$$\dot{\rho}_{ge} = -i\tilde{\omega}_{ge}\rho_{ge} - i\sum_p (V_{gp}\rho_{pe} - \rho_{gp}V_{pe}) \tag{17}$$

At this point we switch to the rotating frame (see Sec. 10.2.2 in [1]), in which the density matrix $\tilde{\rho}$ can be obtained from

$$\tilde{\rho} = U^\dagger \rho U \tag{18}$$

where U is a unitary diagonal matrix given by

$$U_{mm} = \begin{cases} 1 & \text{if } m \text{ is a ground state} \\ e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega t)} & \text{if } m \text{ is an excited state} \end{cases} \tag{19}$$

We thus have $\tilde{\rho}_{ge} = \rho_{ge}e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega t)}$, and the equation of motion for the matrix element in the rotating frame is

$$\begin{aligned}\dot{\tilde{\rho}}_{ge} &= -i\omega\tilde{\rho}_{ge} + \dot{\rho}_{ge}e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega t)} \\ &= -i\omega\tilde{\rho}_{ge} + \left[-i\tilde{\omega}_{ge}\rho_{ge} - i\sum_p (V_{gp}\rho_{pe} - \rho_{gp}V_{pe})\right] e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega t)} \\ &= -i\tilde{\rho}_{ge}\tilde{\omega}'_{ge} - i\sum_p (V_{gp}\rho_{pe} - \rho_{gp}V_{pe})\, e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega t)}\end{aligned} \tag{20}$$

where

$$\tilde{\omega}'_{ge} = \omega + (E_g - E_e)/\hbar - i\Gamma_e/2 \equiv \Delta_{ge} - i\Gamma_e/2 \tag{21}$$

We recall that the interaction term is given by

$$\hbar V = -\boldsymbol{d}\cdot\boldsymbol{\mathcal{E}} = -\boldsymbol{d}\cdot\mathcal{E}_0\mathrm{Re}[\hat{\boldsymbol{\varepsilon}}e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega t)}] = -\boldsymbol{d}\cdot\frac{\mathcal{E}_0(\hat{\boldsymbol{\varepsilon}}e^{i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega t)} + \hat{\boldsymbol{\varepsilon}}^* e^{-i(\boldsymbol{k}\cdot\boldsymbol{r}-i\omega t)})}{2} \tag{22}$$

Introducing this in the equation of motion for $\tilde{\rho}_{ge}$, neglecting the rapidly oscillating terms, and making use of the fact that the wavelength of the light is much larger than the extent of the atom (so that the spatially dependent exponential can be taken outside of matrix elements) one gets to

$$\dot{\tilde{\rho}}_{ge} = -i\tilde{\rho}_{ge}\tilde{\omega}'_{ge} + \frac{i}{2\hbar}\sum_p [\boldsymbol{d}_{gp}\rho_{pe} - \rho_{gp}\boldsymbol{d}_{pe}]\cdot\mathcal{E}_0\hat{\boldsymbol{\varepsilon}}^* \tag{23}$$

which has a steady-state solution given by

$$\tilde{\rho}_{ge} = \frac{1}{2\hbar\tilde{\omega}'_{ge}}\sum_p [\boldsymbol{d}_{gp}\rho_{pe} - \rho_{gp}\boldsymbol{d}_{pe}]\cdot\mathcal{E}_0\hat{\boldsymbol{\varepsilon}}^* \tag{24}$$

Finally, we neglect coherences and populations in the excited states ($\rho_{ee'} = 0$) and coherences in the ground states ($\rho_{gg'} = 0$ if $g \neq g'$). This can be justified since the perturbation is week and is not expected to modify the initial excited state populations (which are zero). Furthermore, coherences between ground states $g \neq g'$ can only be expected if there is population in the excited state. These coherences can only develop is there is excitation from one ground state and then spontaneous or stimulated emission to a different ground state, so they can be neglected in the weak perturbation treatment. The final result is

$$\tilde{\rho}_{ge} = -\frac{\boldsymbol{d}_{ge} \cdot \mathcal{E}_0 \hat{\boldsymbol{\varepsilon}}^*}{\hbar(2\Delta_{ge} - i\Gamma)} \rho_{gg} \tag{25}$$

which after plugging back into the equation for the expectation value of the dipole moment gives

$$\begin{aligned}
\langle \boldsymbol{d} \rangle &= \sum_{ge} 2\mathrm{Re}[\rho_{ge} \boldsymbol{d}_{eg}] \\
&= \sum_{ge} 2\mathrm{Re}[\tilde{\rho}_{ge} e^{-i(\boldsymbol{k}\cdot\boldsymbol{r} - \omega t)} \boldsymbol{d}_{eg}] \\
&= \sum_{ge} \rho_{gg} \mathrm{Re}[-\frac{\boldsymbol{d}_{ge} \cdot \mathcal{E}_0 \hat{\boldsymbol{\varepsilon}}^*}{\hbar(\Delta_{ge} + i\Gamma/2)} e^{-i(\boldsymbol{k}\cdot\boldsymbol{r} - \omega t)} \boldsymbol{d}_{eg}] \\
&\equiv \sum_{ge} \rho_{gg} \mathrm{Re}[-\frac{\boldsymbol{d}_{eg} \cdot \mathcal{E}_0 \hat{\boldsymbol{\varepsilon}}}{\hbar(\Delta_{ge} - i\Gamma/2)} e^{i(\boldsymbol{k}\cdot\boldsymbol{r} - \omega t)} \boldsymbol{d}_{ge}] \\
&= \sum_{ge} \rho_{gg} \mathrm{Re}[-\frac{\boldsymbol{d}_{eg} \cdot \tilde{\boldsymbol{\mathcal{E}}}}{\hbar(\Delta_{ge} - i\Gamma/2)} \boldsymbol{d}_{ge}]
\end{aligned} \tag{26}$$

where in the last line $\tilde{\boldsymbol{\mathcal{E}}} = \mathcal{E}_0 \hat{\boldsymbol{\varepsilon}} e^{i(\boldsymbol{k}\cdot\boldsymbol{r} - \omega t)}$ is the complex electric field vector (not to be confused with the actual electric field vector, which is the real part of it).

Labeling the cartesian components of $\boldsymbol{d}$ with greek upper indices and implying sums over repeated greek indices, we have

$$\begin{aligned}
\langle d^\mu \rangle &= \sum_{ge} \rho_{gg} \mathrm{Re}[-\frac{d^\nu_{eg} \tilde{\mathcal{E}}^\nu d^\mu_{ge}}{\hbar(\Delta_{ge} - i\Gamma/2)}] \\
&= \mathrm{Re}\left[\left(\sum_{ge} -\rho_{gg} \frac{d^\nu_{eg} d^\mu_{ge}}{\hbar(\Delta_{ge} - i\Gamma/2)} \right) \tilde{\mathcal{E}}^\nu \right]
\end{aligned} \tag{27}$$

and from here we can write

$$\mathbf{P} = \mathrm{Re}\left[\epsilon_0 \boldsymbol{\chi} \tilde{\boldsymbol{\mathcal{E}}} \right] \tag{28}$$

where we have defined the electric susceptibility tensor as

$$\begin{aligned}
\chi_{\mu\nu} &= -\frac{n}{\hbar\epsilon_0} \sum_{ge} \rho_{gg} \frac{d^\nu_{eg} d^\mu_{ge}}{\Delta_{ge} - i\Gamma/2} \\
&= -\frac{n}{\hbar\epsilon_0} \sum_{ge} \rho_{gg} \frac{\langle e|d^\nu|g\rangle\langle g|d^\mu|e\rangle}{\Delta_{ge} - i\Gamma/2}
\end{aligned} \tag{29}$$

3 Susceptibility for ^{6}Li atoms at high magnetic field

Going back to the parameterization scheme introduced in Eqs. 1,4 we can identify[1]

$$(P_1 - iP_2)\hat{\boldsymbol{e}}_1 + (P_3 - iP_4)\hat{\boldsymbol{e}}_2 = \epsilon_0\mathcal{E}_0\boldsymbol{\chi}[(\cos\alpha\cos\epsilon - i\sin\alpha\sin\epsilon)\hat{\boldsymbol{e}}_1 + (\sin\alpha\cos\epsilon + i\cos\alpha\sin\epsilon)\hat{\boldsymbol{e}}_2] \quad (30)$$

Using $\hat{\boldsymbol{e}}_1 = \left[\begin{smallmatrix}1\\0\end{smallmatrix}\right]$ and $\hat{\boldsymbol{e}}_2 = \left[\begin{smallmatrix}0\\1\end{smallmatrix}\right]$ this can be written in matrix form as

$$\begin{bmatrix}P_1 - iP_2\\ P_3 - iP_4\end{bmatrix} = \epsilon_0\mathcal{E}_0\begin{bmatrix}\chi_{11} & \chi_{12}\\ \chi_{21} & \chi_{22}\end{bmatrix}\begin{bmatrix}\cos\alpha\cos\epsilon - i\sin\alpha\sin\epsilon\\ \sin\alpha\cos\epsilon + i\cos\alpha\sin\epsilon\end{bmatrix} \quad (31)$$

At this point we have to make a choice for the vectors $\hat{\boldsymbol{e}}_{1,2}$. In our imaging setup the magnetic field is oriented along the z axis, and the probe beam propagate along y, perpendicular to the magnetic field. The polarization of the light lies on the xz plane, so we will asign $\hat{\boldsymbol{e}}_1 = \hat{\boldsymbol{x}}$ and $\hat{\boldsymbol{e}}_2 = \hat{\boldsymbol{z}}$, and we will evaluate the corresponding components of the electric susceptibility tensor: χ_{xx}, χ_{xz}, χ_{zx}, and χ_{zz}.

The first thing to notice is that the off diagonal components of the susceptibility vanish. This is because the x and z components of the dipole cannot couple to thesame excited state. For the diagonal components we will need to evaluate the matrix elements $|\langle e|d_{x,z}|g\rangle|^2$, which can be expressed in terms of the spherical basis components of the dipole moment, d_q, as

$$|\langle e|d_i|g\rangle|^2 = \sum_q |\hat{\boldsymbol{e}}_i \cdot \hat{\boldsymbol{e}}_q|^2 |\langle e|d_q|g\rangle|^2 \quad (32)$$

We recall that the relevant quantum numbers for the ground and excited states are

$$|g\rangle = |R_g\, L\, S\, J\, m_J\rangle \quad (33)$$
$$|e\rangle = |R_e\, L\, S\, J\, m_J\rangle \quad (34)$$
$$(35)$$

At the field of interest, the nuclear spin and its projection are decoupled from the angular momentum of the electron, so they do not play any role in the atomic transition. Figure 1 shows the energy levels of the $2S_{1/2}$ and $2P_{3/2}$ states of a ^{6}Li atom in the presence of a magnetic field. In our experiment we tipically have atoms in an incoherent spin mixure of states $|1\rangle$ and $|2\rangle$. Both of those state have $m_J = -1/2$ so from either of them one can drive transitions to excited states with $m_J = -3/2, -1/2, +1/2$. In what follows we refer to those excited states $|e\rangle$ as $|-\rangle$, $|\pi\rangle$, and $|+\rangle$ respectively.

To evaluate $|\langle e|d_q|g\rangle|^2$, we use the Wigner-Eckart theorem(see Sec. 5.4.1 of [2]) to make the total angularm momentum selection rules explicit in a $3j$-symbol. This defiens the reduced matrix element $|\langle R_e\, L\, S\, J||d||R_g\, L\, S\, J\rangle|^2$,

$$|\langle e|d_q|g\rangle|^2 = \begin{pmatrix}J_e & 1 & J_g\\ -m_{Je} & q & m_{Jg}\end{pmatrix}^2 |\langle R_e\, L\, S\, J||d||R_g\, L\, S\, J\rangle|^2 \quad (36)$$

The dipole moment operator only acts on the electronic angular momentum, L, and our quantum state is expressed in the LS coupled scheme, where L and S are coupled to form $J = L + S$. One can use the formula for the expectation of a single part operator on a coupled scheme (see Sec. 7.1.7 on [2]), in this case the dipole moment operating on the LS coupled state. One obtains

$$|\langle e|d_q|g\rangle|^2 = \begin{pmatrix}J_e & 1 & J_g\\ -m_{Je} & q & m_{Jg}\end{pmatrix}^2 (2J_e+1)(2J_g+1)\begin{Bmatrix}L_g & J_g & S\\ J_e & L_e & 1\end{Bmatrix}^2 |\langle R_e\, L||d||R_g\, L\rangle|^2 \quad (37)$$

[1]The reader is reminded that $\boldsymbol{\chi}$ is a tensor and, for instance, the prodcuct $\boldsymbol{\chi}\hat{\boldsymbol{e}}_1$ may have components along both $\hat{\boldsymbol{e}}_1$ and $\hat{\boldsymbol{e}}_2$.

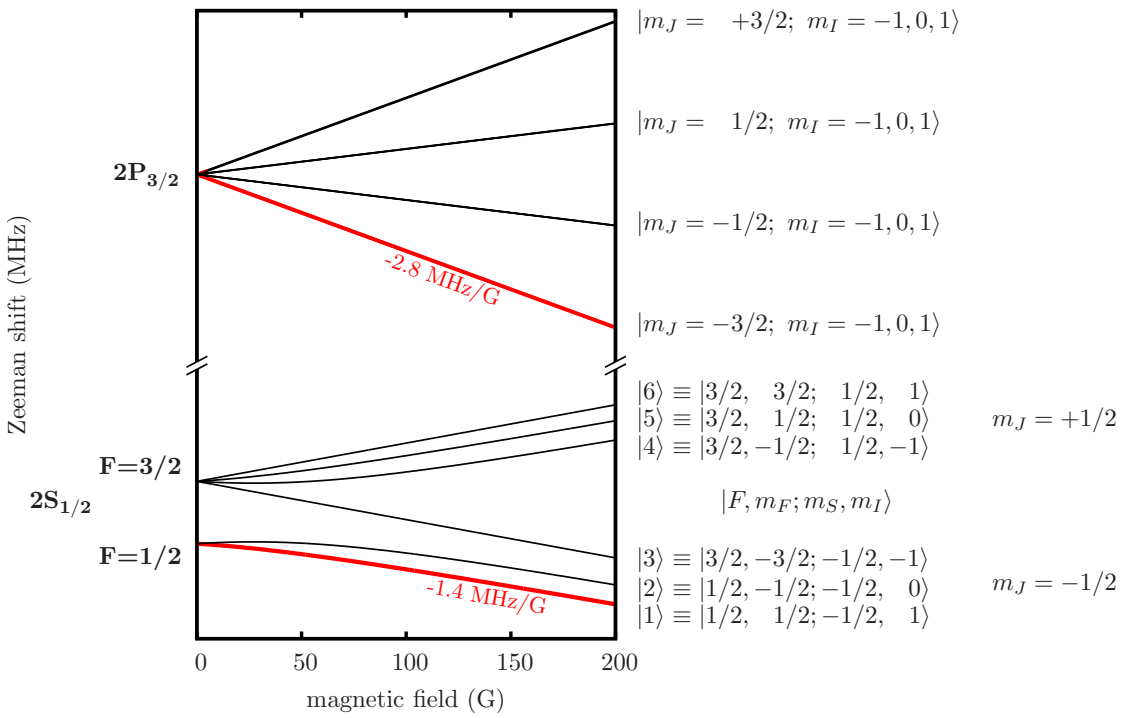

Figure 1: Level diagram relevant for imaging transitions

The fully reduced matrix element can be related to the lifetime, τ, or the natural linewidth, Γ, of the excited state [**?**],

$$|\langle R_e\, L||d||R_g\, L\rangle|^2 = \frac{1}{\tau}\frac{9\epsilon_0\hbar\lambda^3}{8\pi^2} = \Gamma\frac{9\epsilon_0\hbar\lambda^3}{8\pi^2}$$

resulting finally in

$$|\langle e|d_q|g\rangle|^2 = \begin{pmatrix} J_e & 1 & J_g \\ -m_{Je} & q & m_{Jg}\end{pmatrix}^2 (2J_e+1)(2J_g+1)\begin{Bmatrix} L_g & J_g & S \\ J_e & L_e & 1\end{Bmatrix}^2 \Gamma\frac{9\epsilon_0\hbar\lambda^3}{8\pi^2} \tag{38}$$

At this point we plug in some of the values for our case, $L_g = 0$, $J_g = 1/2$, $m_{Jg} = -1/2$, $L_e = 1$, and $J_e = 3/2$. The $6j$ symbol can be evaluated in Mathematica. This results in,

$$|\langle e|d_q|g\rangle|^2 = \begin{pmatrix} J_e & 1 & J_g \\ -m_{Je} & q & m_{Jg}\end{pmatrix}^2 \Gamma\frac{3\epsilon_0\hbar\lambda^3}{2\pi^2} \tag{39}$$

$$|\langle e|d_i|g\rangle|^2 = \sum_q |\hat{\boldsymbol{e}}_i\cdot\hat{\boldsymbol{e}}_q|^2 \begin{pmatrix} J_e & 1 & J_g \\ -m_{Je} & q & m_{Jg}\end{pmatrix}^2 \Gamma\frac{3\epsilon_0\hbar\lambda^3}{2\pi^2} \tag{40}$$

Going back to the x and z components of the dipole moment we have

$$\begin{aligned}
\chi_{xx} &= -\frac{n}{\hbar\epsilon_0}\sum_{ge}\rho_{gg}\frac{|\langle g|d_x|e\rangle|^2}{\Delta_{ge}-i\Gamma/2} \\
&= -\frac{n}{\hbar\epsilon_0}\sum_{g}\rho_{gg}\Gamma\frac{3\epsilon_0\hbar\lambda^3}{2\pi^2}\left(\frac{1}{\Delta_- - i\Gamma/2}|\hat{\boldsymbol{e}}_x\cdot\hat{\boldsymbol{e}}_-|^2\begin{pmatrix}3/2 & 1 & 1/2\\ 3/2 & -1 & -1/2\end{pmatrix}^2\right. \\
&\qquad\qquad \left. + \frac{1}{\Delta_+ - i\Gamma/2}|\hat{\boldsymbol{e}}_x\cdot\hat{\boldsymbol{e}}_+|^2\begin{pmatrix}3/2 & 1 & 1/2\\ -1/2 & +1 & -1/2\end{pmatrix}^2\right) \\
&= -\frac{n}{\hbar\epsilon_0}\sum_{g}\rho_{gg}\Gamma\frac{3\epsilon_0\hbar\lambda^3}{2\pi^2}\left(\frac{1/8}{\Delta_- - i\Gamma/2}+\frac{1/24}{\Delta_+ - i\Gamma/2}\right)
\end{aligned} \tag{41}$$

$$\begin{aligned}
\chi_{zz} &= -\frac{n}{\hbar\epsilon_0}\sum_{ge}\rho_{gg}\frac{|\langle g|d_z|e\rangle|^2}{\Delta_{ge}-i\Gamma/2} \\
&= -\frac{n}{\hbar\epsilon_0}\sum_{g}\rho_{gg}\Gamma\frac{3\epsilon_0\hbar\lambda^3}{2\pi^2}\frac{1}{\Delta_\pi - i\Gamma/2}|\hat{\boldsymbol{e}}_z\cdot\hat{\boldsymbol{e}}_0|^2\begin{pmatrix}3/2 & 1 & 1/2\\ 1/2 & 0 & -1/2\end{pmatrix}^2 \\
&= -\frac{n}{\hbar\epsilon_0}\sum_{g}\rho_{gg}\Gamma\frac{3\epsilon_0\hbar\lambda^3}{2\pi^2}\frac{1/6}{\Delta_\pi - i\Gamma/2}
\end{aligned} \tag{42}$$

In our experiment we have a 50/50 spin mixture of atoms in the ground states $|1\rangle$ and $|2\rangle$, so the sum over ρ_{gg} can be carried out to give

$$\begin{aligned}
\chi_{xx} &= -n\Gamma\frac{3\lambda^3}{2\pi^2}\left(\frac{1/8}{\Delta_- - i\Gamma/2}+\frac{1/24}{\Delta_+ - i\Gamma/2}\right) \\
\chi_{zz} &= -n\Gamma\frac{3\lambda^3}{2\pi^2}\frac{1/6}{\Delta_\pi - i\Gamma/2}
\end{aligned} \tag{43}$$

Returning to the polarization parameters we have

$$\begin{aligned}
P_1 - iP_2 &= \epsilon_0\mathcal{E}_0\chi_{xx}[\cos\alpha\cos\epsilon - i\sin\alpha\sin\epsilon] \\
P_3 - iP_4 &= \epsilon_0\mathcal{E}_0\chi_{zz}[\sin\alpha\cos\epsilon + i\cos\alpha\sin\epsilon]
\end{aligned} \tag{44}$$

which becomes

$$\begin{aligned}
P_1 &= \epsilon_0\mathcal{E}_0[(\cos\alpha\cos\epsilon)\mathrm{Re}\chi_{xx} + (\sin\alpha\sin\epsilon)\mathrm{Im}\chi_{xx}] \\
P_2 &= \epsilon_0\mathcal{E}_0[(\cos\alpha\cos\epsilon)\mathrm{Im}\chi_{xx} + (\sin\alpha\sin\epsilon)\mathrm{Re}\chi_{xx}] \\
P_3 &= \epsilon_0\mathcal{E}_0[(\sin\alpha\cos\epsilon)\mathrm{Re}\chi_{zz} - (\cos\alpha\sin\epsilon)\mathrm{Im}\chi_{zz}] \\
P_4 &= -\epsilon_0\mathcal{E}_0[(\sin\alpha\cos\epsilon)\mathrm{Im}\chi_{zz} + (\cos\alpha\sin\epsilon)\mathrm{Re}\chi_{zz}]
\end{aligned} \tag{45}$$

The ratio of the imaginary to the real part of the susceptibility components goes as $\chi_{ii} \sim \Gamma/\Delta$. In phase-contrast imaging the detuning is chosen so that the imaginary part can be neglected, which results in

$$\begin{aligned}
P_1 &= \epsilon_0\mathcal{E}_0(\cos\alpha\cos\epsilon)\mathrm{Re}\chi_{xx} \\
P_2 &= \epsilon_0\mathcal{E}_0(\sin\alpha\sin\epsilon)\mathrm{Re}\chi_{xx} \\
P_3 &= \epsilon_0\mathcal{E}_0(\sin\alpha\cos\epsilon)\mathrm{Re}\chi_{zz} \\
P_4 &= -\epsilon_0\mathcal{E}_0(\cos\alpha\sin\epsilon)\mathrm{Re}\chi_{zz}
\end{aligned} \tag{46}$$

The differential equations for the electric field parameters, which were obtained in Eq. 9, become

$$\frac{1}{\mathcal{E}_0}\frac{d\mathcal{E}_0}{d\ell} = 0 \tag{47}$$

$$\frac{d\varphi}{d\ell} = \frac{2\pi\omega}{c}\epsilon_0 \sec 2\epsilon[(\cos^2\alpha\cos^2\epsilon - \sin^2\alpha\sin^2\epsilon)\mathrm{Re}\chi_{xx} + (\sin^2\alpha\cos^2\epsilon - \cos^2\alpha\sin^2\epsilon)\mathrm{Re}\chi_{zz}] \tag{48}$$

$$\frac{d\alpha}{d\ell} = \frac{2\pi\omega}{c}\epsilon_0 \sec 2\epsilon[\sin\epsilon\cos\epsilon(\sin^2\alpha - \cos^2\alpha)(\mathrm{Re}\chi_{zz} - \mathrm{Re}\chi_{xx})] \tag{49}$$

$$\frac{d\epsilon}{d\ell} = -\frac{2\pi\omega}{c}\epsilon_0[\cos\alpha\sin\alpha(\mathrm{Re}\chi_{xx} - \mathrm{Re}\chi_{zz})] \tag{50}$$

$$\tag{51}$$

$$\begin{aligned} P_1 - iP_2 &= -\epsilon_0\mathcal{E}_0 n\frac{3\lambda^3\Gamma}{2\pi^2}\left(\frac{1/8}{\Delta_- - i\Gamma/2} + \frac{1/24}{\Delta_+ - i\Gamma/2}\right)[\cos\alpha\cos\epsilon - i\sin\alpha\sin\epsilon] \\ P_3 - iP_4 &= -\epsilon_0\mathcal{E}_0 n\frac{3\lambda^3\Gamma}{2\pi^2}\frac{1/6}{\Delta_\pi - i\Gamma/2}[\sin\alpha\cos\epsilon + i\cos\alpha\sin\epsilon] \end{aligned} \tag{52}$$

which after some manipulation becomes

$$\begin{aligned} P_1 &= \left[\cos\alpha\cos\epsilon\left(\frac{8\Delta_-}{8^2\Delta_-^2 + 4^2\Gamma^2} + \frac{24\Delta_+}{24^2\Delta_+^2 + 12^2\Gamma^2}\right) + \sin\alpha\sin\epsilon\left(\frac{4\Gamma}{8^2\Delta_-^2 + 4^2\Gamma^2} + \frac{12\Gamma}{24^2\Delta_+^2 + 12^2\Gamma^2}\right)\right] \\ P_2 &= \\ P_3 &= \\ P_4 &= \end{aligned} \tag{53}$$

$$\begin{bmatrix} P_1 - iP_2 \\ P_3 - iP_4 \end{bmatrix} = \epsilon_0\mathcal{E}_0 \begin{bmatrix} \chi_{11} & \chi_{12} \\ \chi_{21} & \chi_{22} \end{bmatrix} \begin{bmatrix} \cos\alpha\cos\epsilon - i\sin\alpha\sin\epsilon \\ \sin\alpha\cos\epsilon + i\cos\alpha\sin\epsilon \end{bmatrix} \tag{54}$$

The Liouville equation for the density matrix will be solved by performing a unitary transformation to a rotating frame, as is shown in Chapter 10 of [1], but before we can proceed to define the transformation we must introduce the atomic states of interest in our problem, in other words the basis in which we will represent the atomic density matrix.

The angular monetum projection selection rule for a one photon transition is $\Delta m_J = 0, \pm 1$, which means that we can drive transitions from states $|1\rangle$ and $|2\rangle$ (both of which have $m_J = -1/2$) to the $2P_{3/2}$ excited states with $m_J = -3/2, -1/2, +1/2$. We typically use σ_- polarized light, so we drive transitions to $m_J = -3/2$, which is advantageous because this is a cycling transition, which means it can only decay back to $m_J = -1/2$. In phase-contrast imaging the probe light is detuned far enough from the atomic state that the population in the excited state can in most cases be neglected. The fact that the transition closer to the probe laser is a cycling transition is thus inconseqiential. A basic analysis considers only the $m_J = -3/2$ excited states, but if the detuning is sufficiently large, such as it is sometimes necessary for in-situ imaging of very dense clouds, then we must also take into account the $m_J = -1/2$ and $m_J = +1/2$ states.

The basis set of states thus should consist of eight states. Four of them are state $|1\rangle$, which has a nuclear spin projection $m_I = +1$ plus the three corresponding excited states $m_J = -3/2, -1/2, +1/2$ with $m_I = +1$. The other four correspon to the $m_I = 0$ subspace, which contains $|2\rangle$. Since the electric dipole does not couple the nuclear spins, we will solve the problem in one of the m_I substate, and the solution for the other one will be identical. So the basis set of states that we will consider is $\{|g\rangle, |e_-\rangle, |e_0\rangle, |e_+\rangle\}$, where the first is the ground state and the last three are excited states.

The density matrix is then

$$\boldsymbol{\rho} = \begin{bmatrix} \rho_{gg} & \rho_{+g} & \rho_{0g} & \rho_{-g} \\ \rho_{g+} & \rho_{++} & \rho_{0+} & \rho_{-+} \\ \rho_{g0} & \rho_{+0} & \rho_{0+} & \rho_{-0} \\ \rho_{g-} & \rho_{+-} & \rho_{0-} & \rho_{--} \end{bmatrix} \tag{55}$$

The unperturbed Hamiltonian of the atom is

$$H_0 = \hbar \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & \omega_- & 0 & 0 \\ 0 & 0 & \omega_0 & 0 \\ 0 & 0 & 0 & \omega_+ \end{bmatrix} \tag{56}$$

For the interaction between the atoms and the oscillating electric field of the probe laser, we use the electric dipole Hamiltonian $H_I = -\boldsymbol{d} \cdot \boldsymbol{\mathcal{E}} \cos(\omega t)$, we can expand the vectors in the spherical basis $\{\boldsymbol{e}_\pm, \boldsymbol{e}_0\}$, which results in

$$H_I = -\left(\mathcal{E}_x d_x + \mathcal{E}_y d_y + \mathcal{E}_z d_z\right) \cos(\omega t) \tag{57}$$

To evaluate the matrix for H_I we need to find the matrix elements of the cartesian components of $\boldsymbol{d}$. We evaluate the the components of $\boldsymbol{d}$ in the spherical basis, d_q, where $q \in -1, 0, 1$ and then later we can evaluate the cartesian components by projecting them onto the spherical basis vectors.

To evaluate the matrix elements of the d_q, the Wigner-Eckart theorem[2] is used, in order to make the angular momentum selection rules explicit in the 3-j symbol. This defines the reduced matrix element $\langle e \| d \| g \rangle$,

$$\langle e | d_q | g \rangle = (-1)^{J_e - m_e} \begin{pmatrix} J_e & 1 & J_g \\ -m_{Je} & q & m_{Jg} \end{pmatrix} \langle e \| d \| g \rangle \tag{58}$$

The reduced matrix element satisfies $\langle e \| d \| g \rangle = -\langle g \| d \| e \rangle$, so the reader may verify that (in units of the reduced matrix element)

$$d_{-1} = \frac{\sqrt{3}}{6} \begin{bmatrix} 0 & \sqrt{3} & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ -1 & 0 & 0 & 0 \end{bmatrix} \qquad d_0 = \frac{\sqrt{6}}{6} \begin{bmatrix} 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \qquad d_1 = \frac{\sqrt{3}}{6} \begin{bmatrix} 0 & 0 & 0 & 1 \\ -\sqrt{3} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \tag{59}$$

[2] Sec. 5.4.1 on [2]

and upon using $d_x = \frac{1}{\sqrt{2}}(d_- - d_+)$, $d_y = \frac{i}{\sqrt{2}}(d_- + d_+)$, and $d_z = d_0$ gives

$$d_x = \frac{1}{2\sqrt{6}} \begin{bmatrix} 0 & \sqrt{3} & 0 & -1 \\ \sqrt{3} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ -1 & 0 & 0 & 0 \end{bmatrix} \quad d_y = \frac{i}{2\sqrt{6}} \begin{bmatrix} 0 & \sqrt{3} & 0 & 1 \\ -\sqrt{3} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ -1 & 0 & 0 & 0 \end{bmatrix} \quad d_z = \frac{1}{\sqrt{6}} \begin{bmatrix} 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix} \tag{60}$$

We know define three different Rabi frequencies as

$$\hbar\Omega_x = \frac{\langle e\|d\|g\rangle \mathcal{E}_x}{2\sqrt{6}} \quad \hbar\Omega_y = \frac{\langle e\|d\|g\rangle \mathcal{E}_y}{2\sqrt{6}} \quad \hbar\Omega_z = \frac{\langle e\|d\|g\rangle \mathcal{E}_z}{\sqrt{6}} \tag{61}$$

and we can write

$$H_I = \hbar\cos(\omega t) \begin{bmatrix} 0 & -\sqrt{3}(\Omega_x + i\Omega_y) & -\Omega_z & (\Omega_x - i\Omega_y) \\ -\sqrt{3}(\Omega_x - i\Omega_y) & 0 & 0 & 0 \\ -\Omega_z & 0 & 0 & 0 \\ (\Omega x + i\Omega_y) & 0 & 0 & 0 \end{bmatrix} \tag{62}$$

$|\langle R_e\, L\, S\, J||d||R_g\, L\, S\, J\rangle|^2$,

$$|\langle e|d_q|g\rangle|^2 = \begin{pmatrix} J_e & 1 & J_g \\ -m_{Je} & q & m_{Jg} \end{pmatrix}^2 |\langle R_e\, L\, S\, J||d||R_g\, L\, S\, J\rangle|^2 \tag{63}$$

The dipole moment operator only acts on the electronic angular momentum, L, and our quantum state is expressed in the LS coupled scheme, where L and S are coupled to form $J = L + S$. One can use the formula for the expectation of a single part operator on a coupled scheme[3], in this case the dipole moment operating on the LS coupled state. One obtains

$$|\langle e|d_q|g\rangle|^2 = \begin{pmatrix} J_e & 1 & J_g \\ -m_{Je} & q & m_{Jg} \end{pmatrix}^2 (2J_e+1)(2J_g+1) \begin{Bmatrix} L_g & J_g & S \\ J_e & L_e & 1 \end{Bmatrix}^2 |\langle R_e\, L||d||R_g\, L\rangle|^2 \tag{64}$$

The fully reduced matrix element can be related to the lifetime, τ, or the natural linewidth, Γ, of the excited state [**?**],

$$|\langle R_e\, L||d||R_g\, L\rangle|^2 = \frac{1}{\tau}\frac{9\epsilon_0\hbar\lambda^3}{8\pi^2} = \Gamma\frac{9\epsilon_0\hbar\lambda^3}{8\pi^2}$$

resulting finally in

$$|\langle e|d_q|g\rangle|^2 = \begin{pmatrix} J_e & 1 & J_g \\ -m_{Je} & q & m_{Jg} \end{pmatrix}^2 (2J_e+1)(2J_g+1) \begin{Bmatrix} L_g & J_g & S \\ J_e & L_e & 1 \end{Bmatrix}^2 \Gamma\frac{9\epsilon_0\hbar\lambda^3}{8\pi^2} \tag{65}$$

At this point we plug in some of the values for our case, $L_g = 0$, $J_g = 1/2$, $m_{Jg} = -1/2$, $L_e = 1$, and $J_e = 3/2$. The $6j$ symbol can be evaluated in Mathematica. This results in,

[3] Sec. 7.1.7 on [2]

We perform a transformation to the rotating frame using the unitary matrix U,

where the electron's wave function is: and the equation of motion for the c coefficients is derived from the Schrodinger equation:

$$i\hbar\dot{c}_k(t) = \sum_m c_m(t)e^{-i(E_m-E_k)t/\hbar}\langle k|\hat{H}_{\rm int}(t)|m\rangle \tag{66}$$

The interaction between the atom and the electric field is given by the optical Bloch equations, an $\hat{H}_{\rm int}(t) = -\boldsymbol{d}\cdot\boldsymbol{E}\cos(\omega t)$. It is useful to define the density matrix $\rho_{mn} = c_n^* c_m$, which has the following equation of motion

$$\dot{\rho}_{mn} = \frac{1}{i\hbar}\sum_l \left(\rho_{ln}e^{-i(E_l-E_m)t/\hbar}\langle m|\hat{H_{\rm int}}|l\rangle - \rho_{ml}e^{i(E_l-E_n)t/\hbar}\langle l|\hat{H_{\rm int}}|n\rangle\right) \tag{67}$$

4 Experimental setup for PPCI

References

[1] M. Auzinsh, D. Budker, and S. Rochester, *Optically Polarized Atoms: Understanding Light-atom Interactions* (OUP Oxford, 2010).

[2] A. Edmonds, *Angular Momentum in Quantum Mechanics*, *Investigations in Physics Series* (Princeton University Press, 1996).