ABSTRACT

EXPERIMENTAL DETERMINATION OF THE ELECTRIC QUADRUPOLE MOMENT AND COLLISIONAL DEPOLARIZATION OF J=3/2 CESIUM ATOMS WITH KRYPTON USING LINEAR POLARIZATION SPECTROSCOPY.

by Prakash Koirala

We studied the effect of depolarization of the atomic cesium in the $6p^2P_{3/2}$ level by using two-photon two-color nano-second pulse lasers in collision with krypton buffer gas at various pressures. We used linearly polarized dye lasers at 852.112 nm and at 603.409 nm wavelengths for the $6s^2S_{1/2} \rightarrow 6p^2P_{3/2} \rightarrow 10s^2S_{1/2}$ transition. Excitation of the ground state cesium atoms by linearly polarized light produces an electric quadrupole moment in the excited state. The polarization degree of the emission spectra was measured and was used to calculate the alignment (electric quadrupole moment) in the $6p^2P_{3/2}$ state. Further, the collisional depolarization effect on the linear polarization was measured at various buffer gas pressures from the spectra and it showed a strong pressure dependence on the polarization spectrum. Finally, we extracted the collisional depolarization cross-section of the $6p^2P_{3/2}$ Cs atoms in collision with Kr from the measured linear polarization spectrum.
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Chapter 1

Introduction

Spectroscopic studies deal with the interaction of electromagnetic radiation with matter. The excitation of atoms and molecules by absorbing light quanta leaves the atoms or molecules in an anisotropic state. The excited atoms make a transition back to the ground state with the emission of fluorescence light. Depending on the experimental conditions, excited atoms can be oriented or aligned, and which indicates the anisotropy in the excited state through its angular distribution and polarization.

In the field of atomic collision physics the measurement of polarization has proved to be a good mean for understanding the collision dynamics. Research has constantly been carried out to build up the quantitative understanding about the origin of the emitted light. Due to the advent of lasers, spectroscopy has been widely used to study the light-matter interaction and also in many different fields like photoionization, photodissociation, stimulated raman spectroscopy, polarization spectroscopy, laser cooling and trapping and electromagnetically induced transparency (EIT) [1, 2].

Laser light is monochromatic and in many cases can be tuned to desired wavelength, which has made it a very useful tool for spectroscopy. The high degree of brightness of the laser has opened up new dimensions in two-photon or multi-photon processes leading to dissociation or ionization. The research on investigation of stereo-dynamic effect employs the use of polarized light source. Since the output of laser source, in general, is polarized or can be made so, it’s very useful for the study of stereo-dynamic effects.

Dye lasers pumped by Nd:YAG solid state lasers have been widely used in spectroscopic studies. Dye lasers use organic dyes as the lasing medium which, due to its wide bandwidth, can be tuned to the desired wavelength suitable for any particular excitation of the target sample. Recent researches have proved dye lasers to be very useful to study a single photon or multi-photon excitation that results with the information about the life time of different atomic levels, decay rates and atomic ionization [3].

We studied the linear polarization of the $6s^2S_{1/2} \rightarrow 6p^2P_{3/2} \rightarrow 10s^2S_{1/2}$ transition in $^{133}$Cs atom with krypton buffer gas at pressures ranging from 5 to 100 Torr. The process involves stepwise two-photon two-color on resonance excitation
on atomic $^{133}$Cs. We measured the polarization degree on resonance and also at various krypton buffer gas pressure. The study of depolarization of excited state alkali metals in collision with noble gases results with a wealth of information on relaxation rates of electronic moments and the state multipole dependent depolarization cross-section. The study also benefits numerous practical applications like remotely sensing the composition of planetary atmosphere, fusion plasmas and the interstellar medium. The dynamics of collision of alkali group elements in excited states with noble gas has been studied earlier by using various experimental techniques [4, 5, 6, 7]. Fricke and Lüscher [7] used Cs-lamp and different noble gases at low pressure (up to 30 Torr) and reported collisional cross-section of Cs $6p^2 P_{3/2}$ state. Guiry [4] calculated a collisional depolarization cross-section using both linearly and circularly polarized light radiation in the $6p^2 P_{3/2}$ state Cs-atoms with different noble gases and obtained polarization data at high magnetic fields. Okunevich and Rebane theoretically calculated the disalignment cross-section of Cs in the excited state in presence of different noble gases [8, 9]. In an earlier work, we have studied collisional dynamics between the excited state Cs atoms and ground state Ar atoms [10] from the linear and circular polarization spectra.

In this experiment we used the two-photon, two-color linear polarization spectroscopy technique to measure the alignment and depolarization cross section in the excited state of Cs atoms in collision with ground state Kr atoms.
Chapter 2

General Theory of Polarization Spectra

In this chapter we discuss laser induced fluorescence spectroscopy (section 2.1) briefly followed by the basic physical concept of the terms alignment and orientation (section 2.2). Section 2.3 deals with the formulation of alignment with respect to the interaction geometry applicable for our experiment. We wrap up the chapter with a brief discussion about the excitation scheme and selection rules for our experiment.

2.1 Laser Induced Fluorescence Spectroscopy

The process of spontaneous emission of radiation from excited state atoms due to the absorption of laser photon is called laser induced fluorescence. When an atom is excited by the absorption of a resonant photon it decays back to the lower energy state spontaneously with the release of fluorescence, quite often called laser induced fluorescence (LIF). The measurements of the angular distribution of the polarization of the fluorescence signals gives a clear picture of the dynamics of the process [11],[12],[13].

Fig. 2.1 shows the the general schematic of an experimental set up for LIF spectroscopy. The system comprises mainly the three building blocks - (i) the laser system (ii) the interaction region and (iii) fluorescence detection system. The highly collimated pump and probe laser beams (nanosecond-pulsed excitation in our case) are allowed to enter into the interaction region which is a cesium cell at background pressure of $10^{-4}$ Torr. The fluorescence signal is detected from the observing window by using a photomultiplier tube (PMT) which is sensitive at the fluorescence wavelength. Finally the signal from the detector is processed electronically by using various electronic instrumentation depending upon the nature or requirement of the experimental parameters. We use a boxcar integrator which works best when using nanosecond-pulse excitation.

In our experiment we used pulsed dye laser 1 (pump laser) of wavelength 852.114 nm to excite Cs from $6s^2S_{1/2}$ to $6p^2P_{3/2}$ and dye laser 2 (probe laser) of wavelength 603.409 nm from $6p^2P_{3/2}$ to $10s^2S_{1/2}$ level. Since $10s^2S_{1/2} \rightarrow 6s^2S_{1/2}$
transition is forbidden, we observed the cascade fluorescence from the $9p^2 P_{1/2} \rightarrow 6s^2 S_{1/2}$ transition. The excitation scheme for the electronic transitions under study is depicted in Fig. 2.2.

2.2 Orientation and Alignment

When atoms or molecules are excited by polarized light, it leaves the atoms or molecules with inhomogeneous population in the final states. That is to say the population distribution in different magnetic sub-levels of the excited final state is anisotropic. Fano and Macek [12] for the first time introduced the concept of alignment and orientation to measure the anisotropy of the excited states. The process of inhomogeneous population distribution of the final states with different $|m_J|$ is known as alignment and that for final states with different $\pm m_J$ is the orientation of the atoms.

During the process of light-matter interaction, angular momentum is transferred from the light to the ensemble of atoms. The excited atom in a particular quantum state not only has a given energy $E$ but also has a definite angular momentum $\vec{J}$ quantized in space. For the case of an ensemble of atoms with total angular momentum quantum number $J$ then the $(2J+1)$ (magnetic sub-levels can have $(2J+1)$ values) dimensional vector [13] is

$$\overrightarrow{N} = \begin{pmatrix} N_J \\ N_{J-1} \\ \vdots \\ N_{J+1} \\ N_{-1} \end{pmatrix}$$  \hspace{1cm} (2.1)$$

where $N_m$ is the population in the state $| J, M \rangle$. 

Figure 2.1: Schematic of an experimental set up for Laser Induced Spectroscopy
Figure 2.2: Grotrian diagram for cesium showing the excitation scheme for our experiment.

We can expand this vector as a linear combination of the Cartesian basis vectors as,

$$N = \sum_{L=0}^{(2j+1)} n_L \vec{T}_L$$  \hspace{1cm} (2.2)

where $\vec{T}_L$ are the basis vectors with the property

$$\vec{T}_i \cdot \vec{T}_j = \delta_{ij}$$  \hspace{1cm} (2.3)

where $\delta_{ij}$ is the dirac-delta function and has a value of 1 for $i = j$ and 0 for $i \neq j$ and the co-efficient $n_L = \vec{T} \cdot \vec{N}$ represents $2^L$ multipole moments.

The co-efficient $n_0$ in Eq. 2.2 for $L = 0$ is proportional to the total number of excited atoms in the ensemble and is called monopole component of N. For $L = 1$, $n_1$ is proportional to magnetic dipole moment of the ensemble and is known orientation. For $L = 2$, $n_2$ is proportional to the quadrupole moment of N and is known as alignment.

The orientation and alignment are proportional to

$$\langle P_1(\vec{J} \cdot \hat{z}) \rangle = \frac{\langle J_z \rangle}{|\vec{J}|}$$  \hspace{1cm} (2.4)
and

\[ \langle P_2(\hat{J} \cdot \hat{z}) \rangle = \frac{1}{2} \left( \frac{3J^2 - J^2}{J^2} \right) \]  

(2.5)

respectively.

Figure 2.3: Population versus magnetic sublevels for (a) pure monopole moment (isotropic) (b) alignment and (c) orientation.

Fig. 2.3 represents the form of distribution of atoms in the excited state for a \( J = 2 \) system. If the only moment present is that of the monopole, the magnetic sublevels are equally populated as shown in Fig. 2.3(a). If there is any inhomogeneous population in \( |m_J| \), there is a net alignment (Fig. 2.3(b)). Similarly if the population in the final state is such that it differs for \( +m \) and \( -m \) sub-levels, it is called orientation (Fig. 2.3(c)). In our case, since the atom is excited by a linearly polarized light from the 6s state with \( J=1/2 \) to 6p state with \( J=3/2 \), there is no orientation. The alignment is defined as [13]

\[ \langle A_0 \rangle = \sum_{m'} \frac{|a(m')|^2[3m'^2 - J'(J' + 1)]}{J'(J' + 1)} \]  

(2.6)

For the excitation of Cs atom from 6s^2S_{1/2} state to 6p^2P_{3/2} state with linearly polarized light, where \( \Delta m = 0 \) is the only allowed transition, \( m' \) can take the values \( -1/2 \) and \( 1/2 \) and \( a(m') \) is the Clebsch-Gordon (C-G) coefficient for the transition.
The calculated value for $\langle A_0 \rangle$ is then

$$\langle A_0 \rangle = -\frac{12}{15} = -0.8$$

The relation clearly indicates that $\langle A_0 \rangle$ measures the relative population of small $|m_i|$ states versus large $|m_i|$ states. Hence alignment provides information about the spatial distribution of angular momentum vectors $\vec{J}_i$, which is, in turn related to the shape of the excited state charge distribution. Fig. 2.4 shows the angular variation of the charge distribution as a function of alignment parameter. As it is seen from the figure that for $\langle A_0 \rangle = 0$, the charge distribution is isotropic. If $\langle A_0 \rangle < 0$, the distribution is elongated along the quantization axis and for $\langle A_0 \rangle > 0$, it becomes flattened along the collision axis.

2.3 Alignment Formulation and Interaction Geometry

As already mentioned, the excited state multipole moments (magnetic dipole moment is a measure of orientation, electric quadrupole moment is a measure of alignment) are the measurement of the anisotropy of the excited states. The experimental determination of orientation and alignment of the excited state is carried out by measuring the polarization spectra of the emitted light. Before Fano and Macek introduced the general expression of the intensity of the emitted polarized light owing to an arbitrary excitation process in terms of simple geometrical factors and dynamical parameters like alignment and orientation, scientists had been using the density matrix techniques and Racah algebra along with all the relevant formulae to extract multipole moments from the measured polarization spectra. We briefly discuss here about the basic formulation of the general expression of the intensity of the polarized light in terms of alignment and orientation.
Fig. 2.5 depicts the light-matter interaction geometry for measuring emission polarization. The co-ordinate frames \((x, y, z)\) and \((x', y', z')\) represent the collision frame and detection frame respectively defining the Euler angles \((\phi, \theta, \chi)\) [13]. The \(z\)-axis in the collision frame serves as the incident photon axis and is chosen to be the dominant symmetry axis with \(x\) and \(y\) chosen arbitrarily. For the detection frame \(z'\) is the direction of the observation of emitted photon. Hence the polarization vector lies in \(x' - y'\) plane and can be written as

\[
\hat{\epsilon} = \hat{x'} \cos(\beta) + \hat{y'} \sin(\beta) \tag{2.8}
\]

where \(\beta\) determines the nature of polarization state of the photon to be detected. For \(\beta = 0\) or \(\pi\), the light is linearly polarized along \(x'\) and \(y'\) axis respectively and for \(\beta = \pm \pi\), it’s circularly polarized light.

The intensity of the fluorescence as observed by the detector is given by [13]

\[
I = \frac{1}{3} I_0 \left\{ 1 - \frac{1}{2} h^{(2)}(J_i, J_f) \langle A_0 \rangle^{det} + \frac{3}{2} h^{(2)}(J_i, J_f) \langle A_{2+} \rangle^{det} \cos 2\beta + \frac{3}{2} h^{(1)}(J_i, J_f) \langle O_0 \rangle^{det} \sin 2\beta \right\} \tag{2.9}
\]
where \( h^{(k)}(J_i, J_f) \) \((k = 1 \text{ or } 2)\) are geometrical quantities that depend only on angular momentum quantum numbers of initial and final states. The other quantities \( \langle A_0 \rangle_{\text{det}}, \langle A_{2+} \rangle_{\text{det}} \) and \( \langle O_0 \rangle_{\text{det}} \) are multiple moment operators calculated in the detection frame. These dynamical quantities in the detection frame can be related to those in the collision frame \((x, y, z)\) in terms of Euler angles \((\phi, \theta, \chi)\) as

\[
\langle A_0 \rangle_{\text{det}} = \langle A_0 \rangle P_2(\cos \theta), \quad \langle A_{2+} \rangle_{\text{det}} = \langle A_0 \rangle \left( \frac{1}{2} \sin^2 \theta \cos 2\chi \right)
\]

and

\[
\langle A_{2+} \rangle_{\text{det}} = \langle A_0 \rangle \left( \frac{1}{2} \sin^2 \theta \cos 2\chi \right)
\]

where \( P_2(\cos \theta) = \frac{3}{2} \cos^2 \theta - \frac{1}{2} \) is the second rank Legendre polynomial. \( \langle A_0 \rangle \) and \( \langle O_0 \rangle \) are the expectation values of alignment and orientation in the excited state which can be written following the equations 2.4 and 2.5 as

\[
\langle A_0 \rangle = \frac{\langle 3J_z^2 - J^2 \rangle}{J(J+1)}
\]

and

\[
\langle O_0 \rangle = \frac{\langle J_z \rangle}{\sqrt{J(J+1)}}.
\]

Hence in the collision frame, the Eq. 2.9 becomes

\[
I(\phi, \theta, \chi) = \frac{1}{3} I_0 \left\{ 1 - \frac{1}{2} h^{(2)}(J_i, J_f) \langle A_0 \rangle P_2(\cos \theta) \right. \\
+ \frac{3}{4} h^{(2)}((J_i, J_f) \langle A_0 \rangle \sin^2 \theta \cos 2\chi \cos 2\beta) \\
+ \frac{3}{2} h^{(1)}(J_i, J_f) \langle O_0 \rangle \cos \theta \sin 2\beta \left\} \right.
\]

(2.10)

For linearly polarized light, \( \langle O_0 \rangle \) vanishes and Eq. 2.10 reduces to

\[
I(\phi, \theta, \chi) = \frac{1}{3} I_0 \left\{ 1 - \frac{1}{2} h^{(2)}(J_i, J_f) \langle A_0 \rangle [P_2(\cos \theta) - \frac{3}{2} \sin^2 \theta \cos 2\chi] \right\}
\]

(2.11)

We mounted our detector at \( \theta = \frac{\pi}{2} \) with respect to the collision axis \( z \) and measured the fluorescence intensities at two settings of \( \chi \) when \( \chi = 0, \frac{\pi}{2} \). Let \( I_\parallel \) and \( I_\perp \) be the intensities measured at two different settings at \( \chi = 0, \) and \( \chi = \frac{\pi}{2} \) respectively. From Eq. 2.11 we can re-write the \( I_\parallel \) and \( I_\perp \) as

\[
I_\parallel = I(\phi, \theta = \frac{\pi}{2}, \chi = 0) \\
= \frac{1}{3} I_0 \left\{ 1 - \frac{1}{2} h^{(2)}(J_i, J_f) \langle A_0 \rangle \left[ -\frac{1}{2} - \frac{3}{2} \right] \right\} \\
= \frac{1}{3} I_0 \left\{ 1 + h^{(2)}(J_i, J_f) \langle A_0 \rangle \right\}
\]

(2.12)
and

\[
I_\perp = I(\phi, \theta = \frac{\pi}{2}, \chi = \frac{\pi}{2}) = \frac{1}{3} I_0 \left\{ 1 - \frac{1}{2} h^{(2)}(J_i, J_f) \langle A_0 \rangle \left[ -\frac{1}{2} + \frac{3}{2} \right] \right\} = \frac{1}{3} I_0 \left\{ 1 - \frac{1}{2} h^{(2)}(J_i, J_f) \langle A_0 \rangle \right\}.
\]  

(2.13)

The linear polarization is defined as,

\[
P_L = \frac{(I_\parallel - I_\perp)}{(I_\parallel + I_\perp)}
\]

(2.14)

Eqs. 2.12, 2.13 and 2.14 can be solved to get \( P_L \) in terms of \( \langle A_0 \rangle \) as,

\[
P_L = \frac{3 h^{(2)}(J_i, J_f) \langle A_0 \rangle}{4 + h^{(2)}(J_i, J_f) \langle A_0 \rangle}
\]

(2.15)

The geometrical quantity \( h^{(2)}(J_i, J_f) \) is a known constant that depends upon the angular momentum quantum numbers of the initial and final states \([13]\). For the excitation of cesium atoms from ground state \((6s^{2}S_{1/2})\) to the first excited state \((6p^{2}P_{3/2})\),

\[
h^{(2)}(3/2, 1/2) = \frac{J' + 1}{2J' - 1} = -\frac{5}{4}
\]

and the Eq.2.15 reduces to

\[
P_L = \frac{-15 \langle A_0 \rangle}{16 - 5 \langle A_0 \rangle}.
\]

(2.16)

We determine the linear polarization degree \( P_L \) experimentally and Eq. 2.16 is solved for the value of alignment in the excited state.

### 2.4 Excitation Scheme and Selection Rule

In this section we will discuss the basic properties of cesium followed by the general excitation scheme involved in our experiment and the selection rule.

Cesium is an alkali metal with atomic number 55 and mass number 133 which falls on group 1A of the periodic table. It is highly reactive metal and has low melting point at 28.4°C. It has a simple electronic configuration with one valance electron that makes it easier to formulate the atomic theory calculations. The nuclear spin for \(^{133}\)Cs is 7/2. The ground state \((6s^{2}S_{1/2})\) and first excited state \((6p^{2}P_{3/2})\) split into two hyperfine levels where as the excited state \(6p^{2}P_{3/2}\) level has four hyperfine splitting as given by \( F = I + J......I - J. \) The \(10s^{2}S_{1/2}\) state
Figure 2.6: Cesium $D_1$ and $D_2$ lines with frequency splitting between hyperfine energy levels.

has two hyperfine splittings: $F = I \pm 1/2$. Fig. 2.6 shows the cesium $D_1$ and $D_2$ line with hyperfine splitting of $6p^2P_{1/2}$ and $6p^2P_{3/2}$ levels.

We use a cell that contains pure Cs atoms. Owing to the low melting point of Cs, it can be made vapor even at room temperature. The two equations below describe the relationship between vapor pressure and absolute temperature for solid and liquid phase respectively [14] as

$$\log_{10} P_v = -219.48200 + 1088.676/T - 0.08336185T + 94.88752\log_{10} T, \quad (2.17)$$

$$\log_{10} P_v = 8.22127 - 4006.048/T - 0.00060194T - 0.19623\log_{10} T \quad (2.18)$$

where $P_v$ is the vapor pressure in Torr and $T$ is the temperature in Kelvin. The curve of vapor pressure versus temperature from the model of Eq. 2.17 is shown in Fig. 2.7.

Fig. 2.2 shows the general excitation scheme of the cesium atoms in our experiment. The ground state cesium atoms are excited to the intermediate state $6p^2P_{3/2}$ with the absorption of a photon of suitable wavelength. The excited Cs atoms are further pumped up to the $10s^2S_{1/2}$ level by the absorption of another photon of suitable wavelength. In our experiment we used two-photon two-color
nano-second pulsed lasers for the excitation process. The dye laser 1 (pump laser) which matches the wavelength for the transition excites the cesium atoms from ground state to the 6p^2P_{3/2} state and dye laser 2 finally takes the excited cesium atoms from 6p^2P_{3/2} level to 10s^2S_{1/2} level. Since S-S transitions are dipole forbidden, we observed the cascade fluorescence from 9p^2P_{1/2} to 6s^2S_{1/2} level.

Absorption of a photon with suitable wavelength by an atom results in the excitation of the atom in higher energy level. Any excited energy level has a definite value of its angular momentum \( \vec{J} \) that determines the number of magnetic sublevels for that energy level. The state of polarization of light is crucial to determine the magnetic sublevel that will be populated during the excitation process. An atom excited by a linearly polarized light results with a change in magnetic quantum number by zero, i.e \( \Delta m = 0 \). For the excitation by right circularly and left circularly polarized light the transition is such that the change in magnetic quantum number \( \Delta m = +1 \) and \( \Delta m = -1 \), respectively. Selection rules describe the change that takes place in quantum numbers during the atomic transition. It rules out the possibility of transition between all possible pairs of energy levels. Those transitions which are not allowed by selection rule are forbidden. Hence this is the rule that determines the possible transitions among the quantum levels with the emission or absorption of radiation.
Fig. 2.8 shows the different energy levels with possible magnetic sublevels and allowed transitions involved in our experiment. The ground state $6s^2S_{1/2}$ has two magnetic sublevels corresponding to $m = +1/2$ and $m = -1/2$. The $6p^2P_{3/2}$ level has four; $m = +3/2, +1/2, -1/2, -3/2$ and finally the $10s^2S_{1/2}$ state has two sublevels; $m = +1/2, -1/2$. For the excitation of the atoms with linearly polarized radiation along $z$-axis, $\Delta m = 0$ when the direction of polarization of both the lasers are parallel. If the direction of polarization of the lasers are perpendicular to each other, the possible transitions are such that the change in magnetic quantum numbers $\Delta m = \pm 1$. 
Chapter 3

Experimental Set-ups

In this chapter we discuss in detail the experimental apparatus used to measure the polarization data. The section 3.1 of this chapter covers the fundamentals of the Nd:YAG laser followed by the construction details of dye lasers used in the experiment. In section 3.2 we briefly discuss the spectral distribution and longitudinal modes of the dye laser cavities. Section 3.3 presents the implementation of liquid crystal variable retarder and Glan-Thomson polarizer. In the sections 3.4 and 3.5 we discuss the cesium cell, the construction of the vacuum system and the signal detection scheme.

3.1 Lasers

3.1.1 Nd:YAG Laser

Nowadays fixed-wavelength lasers are widely used as pumping light source for tunable dye laser systems. Due to the low efficiency of fixed-wavelength gas lasers in the context of conversion of electrical energy into laser light, solid state lasers have widely been used in many scientific applications. Neodymium atoms (Nd) doped in an ytrrium aluminium garnet crystal, \( Y_3Al_5O_{12} \), abbreviated as Nd:YAG is a most commonly used solid state laser material. The doping results with Nd\(^{3+}\) ion which forms lasing medium. Fig. 3.1 shows the schematic of energy level diagram for the lasing action.

The ground state \( ^4I_{9/2} \) neodymium ions (Nd\(^{3+}\)) are excited to the \( ^4F_{3/2} \) energy level with light of suitable wavelength within the range of 800-820 nm by Xe flash lamp or by 808 nm GaAlAs diode lasers. The excited ions make non-radiative transition to the \( ^4F_{3/2} \) state which has a lifetime of nearly 230 \( \mu \)s. The laser transition occurs between the energy levels \( ^4F_{3/2} \) and \( ^4I_{11/2} \) corresponding to a wavelength \( \lambda = 1064.13 \) nm. The ions finally jumps to the ground state through vibrational relaxation. The laser radiation at \( \lambda = 1064 \) nm can easily be converted to achieve second and third harmonic generation.

In our experiment, we use Continuum Surelite Nd:YAG pulse laser operating at the second harmonic generation to produce 532 nm wavelength with average power of 4.5 W having (4-6) ns pulse width and 20 Hz repetition frequency. This
Figure 3.1: Energy level scheme for Nd:YAG laser.

laser is the pumping source for our dye lasers. Three beam splitters in the path of YAG laser beam splits the beam before the beam is directed to the dye laser apparatus. Each of the beam splitters are anti-reflective (AR) coated on one side (CVI W1-PW1-1012-C-532-45UNP) so as to prevent ghost reflections and allow approximately 9% of the YAG laser power to pass in. The average power of YAG laser as measured by a power meter (Molelectron Detector Inc., PowerMax PM10V1) right after the first and second splitters is 0.2 W and 0.18 W respectively.

3.1.2 Dye Lasers

Tunable dye laser systems basically consist of different gain media like dyes in solution, doped solid state crystals and light-emitting semiconductor diode materials. Dye laser, as indicated by its name, uses organic dyes as gain medium which can be pumped by radiation from a laser source or even by incoherent light source. We use 532 nm pulsed Nd:YAG laser as a pumping source for our home-built dye lasers. Fig. 3.2 represents a sketch for pulsed dye laser resonator set-up for Litmann-Metcalf configuration based on grazing incidence grating.

As mentioned earlier, the dye laser 1 excites Cs atoms from ground state \((6s^2S_{1/2})\) to intermediate state \((6p^2P_{3/2})\). For the generation of the dye laser 1 we use an organic dye LDS 867 (Exciton Inc.) diluted with methanol with the concentration of 84 mg/l. Since the dye degrades very fast, it is constantly circulated into the cuvette (NSG Precision Cells Inc., T-524) of 2 ml capacity by using a dye circulator (Spectra Physics Model 376) to maintain the constant output power of
Figure 3.2: Littman-Metcalf Configuration for tunable dye laser.

The dye laser. A gold-coated grating (Edmund Industrial Optics, Y55-261) having dimension of 30mm x 30mm with 1200 grooves/mm is placed in the oscillation cavity as shown in Fig. 3.2. The output-coupler (CV1 LW-2-1037-C) reflects few percent of the light back into the cavity. The YAG laser is focused at the edge of the dye cell by using cylindrical lens having focal length of 5.08cm. The tuning mirror (Thorlab Inc., BB1-E03) has 99.0% reflectivity in the 750-1150nm range that can be rotated using high precision ultrastable mirror mount to get desired wavelength. The dye laser 1 has a peak wavelength at 853.112nm and can be tuned in the wavelength range of 830-920nm.

For the production of dye laser 2 that excites Cs atoms from intermediate $6p^2P_{3/2}$ state to the final $10s^2S_{1/2}$ state we use rhodamine 640 perchlorate (Exciton Inc.) diluted with methanal. The concentration level of the dye solution of oscillator and amplifier (that amplifies laser 2 by a factor of 9) are 141.8 mg/l and 18.9 mg/l as prescribed by Exciton catalog (www.exciton.com). The dye cells used for both oscillator and amplifier are anti-reflective (AR) coated quartz of 3.2 ml capacity and 8 mm light path length. A holographic grating (Edmund Industrial Optics, Y43-215) having 1200 grooves/mm with 12.5mm x25mm dimension is used in the cavity of laser 2. A broadband mirror (Thorlabs Inc., BB1-E02) with 99.7% reflectivity in the range of 400-900 nm wavelength is used as a tuning mirror. The cylindrical lens and output coupler are identical to those used in laser 1 cavity. The tuning mirror is mounted on a high precision ultra-stable kinematic mirror mount (Newport 610 Series) which has a coarse and fine adjustment for vertical and horizontal axes. The horizontal fine scale is driven by a stepper mother driver.
(Ardel Kinematics Corp, Motor Mike) which is remotely controlled by a computer to tune the wavelength. We use wave meter (Coherent Wave Master) to measure the wavelength of our lasers with an accuracy of 0.001 nm within the range of 380 nm to 1095 nm. In order to view laser 1 so that we can align it, we use infrared viewer (Electrophysics, Electroviewer 7215) or infrared card (Thorlabs Inc., IRC3). We use irises after the output coupler of pump laser and probe laser amplifier to eliminate the unwanted light and pick up the center of the laser beams. We use different converging lenses combination to get the highly collimated beam so that the lasers overlap at the interaction region. The pumping scheme for the dye lasers is shown in Fig. 3.8.

### 3.2 Longitudinal Modes & Free Spectral Range of Dye Laser Cavities

![Standing wave patterns](image)

Figure 3.3: Standing wave patterns within the laser resonator for \( n = 1, 2, 3 \) and 4.

The waves confined to an optical resonator form standing waves pattern in such a way that the length of the resonator is integral multiple of half of their wavelengths. Laser resonator are normally much longer than wide, we consider the longitudinal components with respect to the optical axis of the resonator. If \( L \) be the length of the laser resonator and \( \lambda_n \) be the resonance wavelengths, we can write

\[
L = n \frac{\lambda_n}{2}
\]

\[
= n \frac{c}{2\nu_n}
\]

(3.1)

where \( \nu_n \) is the resonance frequencies and \( c \) is the speed of light. Rearranging the
Eq. 3.1 we get,

\[ \nu_n = \left( \frac{n c}{2L} \right) \]  \hspace{1cm} (3.2)

The mode spacing between two neighboring modes \( \nu_n \) and \( \nu_{n-1} \) is called free spectral range (FSR) for the given resonator.

Hence,

\[
FSR = \Delta \nu \\
= (\nu_n - \nu_{n-1}) \\
= \frac{c}{2L} .
\]  \hspace{1cm} (3.3)

The resonating lengths for laser 1 and laser 2 are 12cm and 11.5cm respectively. The calculated value of FSR for laser 1 is 1.25GHz and for laser 2 is 1.30GHz.

![Figure 3.4: Polarization states of dye lasers in the interaction region.](image_url)

\[ \text{For } I_{\parallel, \parallel} \quad \theta = 0^\circ \]
\[ \text{For } I_{\perp, \perp} \quad \theta = 90^\circ \]

\[ 3.3 \quad \text{Glan-Thompson Polarizer & Liquid Crystal Variable Retarder} \]

A pair of Glan-Thompson Polarizers (GT) (Thorlabs Inc., GTH10M) are positioned in the path of dye lasers just before laser lights enter into the cesium cell. Despite the fact that the YAG laser is vertically polarized along the laboratory z-axis (quantization axis) and dye laser resonators do not change the nature of polarization, GT polarizers ensure the higher degree of polarization which is the requirement for our experiment. We make sure that the laser beams are perfectly aligned and they overlap at the center of cesium cell. Laser 1 and laser 2 collectively excite the cesium atoms to the \( 10s^2S_{1/2} \) level. We intend to measure the intensities of emission spectrum (a) when the plane of polarization of laser 1 and laser 2 are parallel to each other (denoted by \( I_{||} \)) and (b) when they are perpendicular to
each other (denoted by $I_\perp$). We keep the polarization state of dye laser 1 fixed (along laboratory z-axis) and alternately change the plane of polarization of dye laser 2 between $\pi/2$ and 0 by using a Liquid Crystal Variable Retarder (LCR) (Meadowlark Optics, D3040) right after the GTP in the path of laser 2 before the laser enters into the cesium cell. LCR consists nematic crystal that changes the polarization of light wave as it passed through the crystal. The intensity of applied electric field determines the extent of the change in polarization. The potential applied to the cavity of LCR is remotely controlled by using computer. We can change the plane of polarization of dye laser 2 by $\pi/2$ with respect to that of dye laser 1 by applying about 1.5 V and make it parallel by applying 6 V. The LCR is mounted on a high precision rotation mount (Thorlabs Inc., PRM1) to achieve high level of precision. Fig. 3.4 shows polarization states of the dye lasers in the interaction region.

Contrast ratio measures how effectively the LCR changes the polarization state of the light as it passes through it. We require to make the plane of polarization of the dye lasers parallel and perpendicular to each other in an alternate fashion. An application of around 6 V of potential to the LCR makes the plane of polarization of laser 2 parallel and around 1.5 V makes it perpendicular to the plane of polarization of laser 1. The potential is remotely controlled by a computer driver software program (Meadowlark Optics, Cell Drive 3000). To measure the CR, we place a photodiode right after the GTP 1 which is set to polarize the laser 1. But this time we block the laser 1. The photodiode is connected to the oscilloscope. The dye laser 1 passes through the GTP 2, LCR, oven and finally through GTP1 before it enters into the photo diode (Fig. 3.5). To prevent the saturation of the photo diode, we use different neutral density filters that attenuates the power of dye laser 2. We adjust the voltage of LCR so that the plane polarization of dye laser 2 is parallel to the the polarizing plane of GTP 1 so that the maximum intensity is observed in the oscilloscope and we note it down. There after we change the voltage to some other value so that the intensity of the signal on the oscilloscope drops down significantly. Fine adjustments are done by rotating the LCR with the high precision mount to obtain hight degree of accuracy. The ratio of minimum to
maximum intensities is the contrast ratio of LCR. We calculate the contrast ratio on daily basis before we start taking data for the day. We intend to make this ratio really low that ensures the required operation of LCR.

## 3.4 Cesium Cell and Vacuum System

The cesium cell we use in our experiment is a small tubing made up of pyrex with the length of 5.08cm and 2.54cm diameter that contains cesium atoms at extremely low pressure. The cell is kept inside an aluminium oven. The oven is a cylinder with two windows in either side so that the lasers can pass into the cell. The diameter of each of the windows is 1.27 cm. The laser beams overlap inside the cesium cell and the fluorescence signal comes out from a third window in the oven. A concave mirror placed inside the oven at the back of the observation window helps to collect more fluorescence signals. The oven is heated with a flexible flat heavy insulated electric heating tape (Cole Parmer, 36050-20) wrapped around the oven. The oven is further covered with fiber flax and aluminium to ensure the uniform temperature inside the cell. A thermocouple (Omega, KMQSS-020U) attached to the cell monitors the temperature inside the oven. The heating tape and the thermocouple are connected to a temperature controller (Cole Parmer, Digi-Sense EW-89000-00) that regulates the temperature of the cell with the temperature sensitivity of ±0.1°C. The cell is connected to the vacuum chamber via a narrow stem protruding out form the cell as shown in Fig. 3.6 with two gate valves GV 1 and GV 2. A cylinder containing high pressure krypton gas (PRAXAIR, PQL # 922-5129-50) is also connected to the vacuum chamber. A mechanical pump sucks the air out of the chamber fairly below the atmospheric pressure which is followed by a Turbo pump (Varian, Turbo-V60) that reduces the pressure up to $10^{-6}$ Torr.
A digital pressure gauge (HPS, Piezo Display Module) constantly monitors the pressure in the chamber. Closing the valves GV 1 and GV 5 and opening all others, the air in the chamber is pumped out so that the pressure in the chamber drops down to $10^{-6}$ Torr. We then open the gate valve 5 and let the gas expand within the region between GV 4 and GV 5. By opening valves 1 and 2 and closing 3, we slowly open the valve 4 so that the pressure gauge reads nearly the pressure we wish to have in the cesium cell. Finally we open the valve 1 and allow the gas enter into the cesium cell. This might cause the pressure in the chamber drop slightly as the expand in the cesium cell. If the pressure is lower than what we wish to have, we gently open the valve 4 followed by a quick closing. If we feel that the pressure in the chamber before we allow the gas pass into the cesium cell is high enough and we don’t think that we can have a desired pressure in the cell, the gas is pumped out by opening the valve 3. In this way we can set the desired pressure of the buffer gas in our cell.

### 3.5 Signal Detection & Data Processing Set-up

Once the cesium atoms in the cell are excited to $10s^2S_{1/2}$ state, a non radiating transition takes place between this state and $9p^2P_{1/2}$ state. Finally we detect a fluorescence signal of wavelength 361.730 nm when the atoms in $9p^2P_{1/2}$ state make a transition to the ground state. This signal coming out of the oven enters into a photo multiplier tube (PMT) (Hamamatsu, R955). The fluorescence signal hits the photo cathode of the PMT and produces a large number of photo electrons. These electrons are further multiplied by secondary emission. The number of electrons so produced is proportional to the intensity of fluorescence signal that enters into the tube. The PMT works under the application of negative potential of about 900 volt by using high voltage power supply (Stanford Research Inc, PS 350). We use a combination of interference filter (Coherent, 35-3045-000) and colored-glass filter (Coherent, UG11) to detect the signal. The interference filter allows 31% of the desired wavelength and has peak transmission wavelength at 366.7 nm at FWHM of 10.7 nm. The colored-glass filter helps to eliminate the scattered YAG and dye lasers. The output signal from the PMT is connected to a two-stage amplifier (Stanford Research Inc., SR240) that amplifies the signal by 5 times in each stage. The amplified signal then goes to the boxcar averager/integrator (Stanford Research Inc., SR250). The averager is triggered externally by dye laser 1 picked up by a photo diode. We set the gate width of the boxcar averager to 60 ns so that the signal in the oscilloscope is within the range of this time scale. The setting of the gate width also helps to reduce the noise in the signal. The voltage sensitivity in the averager is adjusted according to the input signal strength. The averaged output signal is sent to the Data Acquisition Board (DAQ)(National Instruments, PCI6014) through an outbreak box (National Instruments, CB-68LP). The DAQ board converts the analog signal to digital signal which is further processed by LabVIEW program (National Instruments, LabVIEW 7.0). By using this program we can have the control over the polarization and frequency scan of the dye laser 2. We use micrometer sub VI.vi program to change the wavelength of dye laser.
2 until we get the resonance wavelength which is indicated by large signal in the oscilloscope. Finally we take the linear polarization data at resonance by using `takedata2_sub.vi` program which is the average of 1500 data points. By using the Eq. 2.14 we use the polarization data so obtained to calculate the polarization degree for the emission spectra. The diagram in Fig. 3.7 shows the set-up for signal detection scheme in detail. The Table 3.1 lists the different kinds of softwares we used in our experiment.

Table 3.1: List of the software used in our experiment.

<table>
<thead>
<tr>
<th>S.N.</th>
<th>Task</th>
<th>Software</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>LCR Voltage Application</td>
<td>CellDRIVE 3000 Advanced.exe</td>
</tr>
<tr>
<td>2.</td>
<td>Scanning for resonance signal</td>
<td>Get Spectrum Test.vi</td>
</tr>
<tr>
<td>3.</td>
<td>Tuning for laser 2</td>
<td>micrometer subVI2.vi</td>
</tr>
<tr>
<td>4.</td>
<td>Taking Data</td>
<td>takedata2_sub.vi</td>
</tr>
</tbody>
</table>
Figure 3.8: Pumping scheme for dye lasers.
Figure 3.9: Experimental apparatus.
Chapter 4

Experimental Results

In this chapter we discuss the experimental measurement overview briefly in section 4.1 followed by systematic effects that includes magnetic field effect, spectral broadening mechanism, laser power effect and temperature effect on the measured linear polarization spectrum in section 4.2. In section 4.3 we have calculated the alignment in $6s^2P_{3/2}$ level. The last two sections 4.4 and 4.5 deal with the depolarization of the linear polarization in presence of krypton buffer gas and extraction of depolarization cross-section from the experimentally determined parameters.

4.1 Overview of Measurement

The polarization spectrum for the $6s^2S_{1/2} \rightarrow 6p^2P_{1/2} \rightarrow 10s^2S_{1/2}$ transition was measured by using a two-color two-photon excitation process in atomic cesium. We used 530 nm Nd:YAG laser to pump the home-built dye lasers. We set the wavelength of dye laser 1 at 852.112 nm and the wavelength of dye laser 2 at 603.409 nm. Owing to the low power of dye laser 1, we could not measure its wavelength by using wave meter. For the confirmation of right wavelength of laser 1 we observed Cs D2 signal by using a combination of interference filter and color glass filter. By changing the micrometer scale attached to the dye laser 1 cavity, we obtained the peak signal in the oscilloscope which corresponds to the wavelength of 852.112 nm of laser 1. After setting the laser 1 at resonance, we changed the interference filter for two photon transition and observed the two photon signal. For the reduction of noise, we covered the housing of PMT by black clothes which prevented the scattered light from going into the PMT. For the same reason we covered the signal wire with a copper braided wire. We passed the dye lasers through the polarizers before they entered into the cell. The plane of polarization of our dye laser 2 could be changed to make it parallel and perpendicular to dye laser 1 by applying 6 volt and about 1.5 volt to the remotely controlled LCVR. By tuning the wavelengths of lasers we got two photon signal. The gate width of the Boxcar averager/integrator was adjusted so that the signal could be averaged within the width. The fluorescence signal was amplified 25 times before it was connected to the oscilloscope. By using takedata3_sub.vi program we measured the signal. We adjusted the sensitivity of Boxcar averager/integrator in accordance with the
Table 4.1: Important parameters used in the experiment.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_{Y_{AG}}$</td>
<td>1.85 W</td>
</tr>
<tr>
<td>$P_{L_1}$</td>
<td>0.5 W</td>
</tr>
<tr>
<td>$P_{L_2}$</td>
<td>5 W</td>
</tr>
<tr>
<td>$6s^2S_{1/2} \rightarrow 6p^2P_{3/2}$</td>
<td>852.112nm</td>
</tr>
<tr>
<td>$6p^2P_{3/2} \rightarrow 10s^2S_{1/2}$</td>
<td>603.409nm</td>
</tr>
<tr>
<td>$9p^2P_{3/2} \rightarrow 6s^2S_{1/2}$</td>
<td>361.730nm</td>
</tr>
<tr>
<td>Dye concentration for L1</td>
<td>84mg/l(methanol)</td>
</tr>
<tr>
<td>Dye concentration for L2 oscillator</td>
<td>114mg/l(methanol)</td>
</tr>
<tr>
<td>Dye concentration for L2 amplifier</td>
<td>16.83mg/l(methanol)</td>
</tr>
<tr>
<td>Gatewidth of the signal</td>
<td>60 ns</td>
</tr>
</tbody>
</table>

signal strength of our fluorescence. For larger signal strength’ it requires larger value of the sensitivity for better result. We opened the *takedata3_sub.vi* program and provided the base line by blocking the laser 1. This helped to ignore the noise that could have somehow found its way into the signal. The power of lasers and contrast ratio was measured in daily basis. If the power of lasers had dropped down, the dyes were refreshed. Once we made sure that the contrast ratio was good, we started taking the polarization data. Each data was an average of 1500 data points. The data so obtained was analyzed by using OriginPro7 program. Table 4.1 shows some important parameters involved in our experiment.

### 4.2 Systematic Effects

Systematic effects that might bring a change in the polarization measurement are mainly due to the temperature of the cell, power of the lasers, magnetic field effect and hyperfine splitting. By using pump-probe technique we can reduce the hyperfine depolarization effect. In our research we don’t see significant temperature dependence on the measured polarization measurement. Besides these effects we may have birefringence of oven window and imperfection of polarizer which cause to change the direction and uniformity of our laser. We did our best to maintain the direction and uniformity of our laser. The GTP produces the pure linearly polarized light. The extinction ration of the polarizers is checked to ensure the purity of polarization of our lasers. Due attention was paid to have linear, uniform and perfectly aligned laser beams.

#### 4.2.1 B-field Effect

If an atom is immersed in an external magnetic field, the atomic magnetic moment of the atom interacts with the external magnetic field. This interaction will split the energy levels of the atom. The splitting spectral lines in magnetic field is called Zeeman effect. The energy shift of an energy level in a magnetic field is given by
\[ U = g m_j \frac{e \hbar B}{2 m_e} \] \hspace{1cm} (4.1)

where \( g \) is called the Lande g-factor, \( m_j \) is the magnetic quantum number, \( B \) is the strength of external magnetic field and \( m_e \) is the electronic mass. The Lande g-factor is given by,

\[ g = 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)} \] \hspace{1cm} (4.2)

For Cs \( 6p^2P_{3/2} \) level, the calculated value for \( g \) is 4/3. Hence the energy shift for the levels \( m_{3/2} \) and \( m_{-3/2} \) is given by,

\[ U = \pm \frac{e \hbar B}{m_e} \] \hspace{1cm} (4.3)

So the maximum possible energy shift for \( 6p^2P_{3/2} \) level is given by,

\[ \Delta U = 2 \frac{e \hbar B}{m_e} \] \hspace{1cm} (4.4)

The frequency corresponding to this splitting is

\[ \nu = \frac{e B}{\pi m_e} \] \hspace{1cm} (4.5)

The earth’s magnetic field has a maximum value of 0.6 gauss at the magnetic poles and smaller elsewhere. We use heating tape (Cole Parmer, EW-36050-20) working at 468 watts, 120 VAC. Hence the maximum current drawn by the tape comes out to be 3.9 A. The magnetic field produced by this tape at the axis of our cesium cell has a maximum value of 0.1 gauss. The current in the coil was passed in such a way that the magnetic field produced by this heating tape was directed opposite to the direction of earth’s magnetic field. Hence the maximum possible total external magnetic field strength inside the interaction region is about 0.40 gauss. Submitting this value in Eq. 4.5, we get the frequency corresponding to the maximum possible splitting of the \( 6p^2P_{3/2} \) level. We calculated it to be 2.23 MHz. Proceeding in the same way the Zeeman splitting for \( 6s^2S_{1/2} \) level is 1.11 MHz. The bandwidths of our dye lasers are much more larger than the frequency splitting for these levels due to residual magnetic field. So the effect of external magnetic field in our experiment has no contribution to alter the polarization degree.

### 4.2.2 Broadening Mechanism of Spectral lines

There are different kinds of broadening mechanism in the spectral lines. The lifetime for \( 6p^2P_{3/2} \) state is 30.67 ns. The uncertainty principle suggests that for the particles with extremely short lifetimes, the uncertainty in its energy is significantly high. Hence the major source of broadening for cesium \( 6p^2P_{3/2} \) level is the natural linewidth which is about 5 MHz at full width half maxima. The spectral line may further broaden due to other broadening mechanisms such as pressure broadening,
wall-collision broadening, Doppler broadening etc. The atoms in a gas move around randomly and collide with each other and the walls of the container. This process shortens the lifetime of the excited states and gives additional broadening to the spectral lines. The average time for collision between two cesium atoms can be written as,

$$\tau_c = \frac{1}{4\sigma_{Cs-Cs}P} \left( \frac{mk_BT}{3} \right)^{1/2}$$  \hspace{1cm} (4.6)

where \(P\) is the pressure, \(T\) is the absolute temperature, \(m\) is the mass of cesium atom, \(k_B\) is the Boltzmann’s constant and \(\sigma_{Cs-Cs}\) is the collision cross-section which can be calculated by using the following relation.

$$\sigma_{Cs-Cs} = \pi r^2$$  \hspace{1cm} (4.7)

where \(r\) is the radius of the cesium atom.

The corresponding bandwidth at full width half maxima is given by,

$$\Delta \nu = \frac{1}{2\pi \tau_c}$$  \hspace{1cm} (4.8)

By using the equations above we calculated the bandwidth at the pressure of 1 Torr and temperature of 363.16 K to be 5.52 MHz. The numerical values of the physical parameters involved in the calculation is listed in Table 4.2. We used pure cesium cell at the background pressure of 10^{-4} Torr for our experiment and the calculated value of line width due to the collision between the cesium atoms at the temperature of 363.16 K was 522 Hz which is very small as compared with the natural line width of the cesium 6p2 \(P_{3/2}\) state. Hence collision broadening has no effect to change the polarization degree of the spectrum.

Similarly, the collision of the excited atoms with the wall of the containing vessel may reduce the lifetime in excited state resulting with further broadening of the spectral line. This broadening is directly proportional to the average speed of the atoms which is the function of temperature and inversely proportional to the length of the container. We used cesium cell having length of about 5.08 cm. The wall-collision rate may be written as [16],

$$\Delta \nu_w = \frac{v_{Cs}}{2L}$$  \hspace{1cm} (4.9)

where \(v_{Cs}\) is the velocity of cesium atom at given temperature which can be calculated by using the relation,

$$\frac{1}{2}mv_{Cs}^2 = \frac{1}{2}k_BT$$  \hspace{1cm} (4.10)

where \(k_B\) is the Boltzmann’s constant and \(T\) is the absolute temperature.

The calculated value of the velocity of cesium atoms at the temperature of 363.16 K was 150.185 m/s. Submitting this value in Eq. 4.9 we get the line width due to wall-collision to be 1.416 KHz which is very very small in comparison with natural bandwidth as well as Cs-Cs collision line width. Therefore the we conclude that the wall-collision of cesium atoms has no significant effect to broaden the spectral lines and alter linear polarization degree.
Table 4.2: Numerical values for the physical constants used in bandwidth calculation.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
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</thead>
<tbody>
<tr>
<td>$m$</td>
<td>$2.223 \times 10^{-25} \text{kg}$</td>
</tr>
<tr>
<td>$k_B$</td>
<td>$1.3807 \times 10^{-23} \text{JK}^{-1}$</td>
</tr>
<tr>
<td>$r$</td>
<td>$6.3 \times 10^{-10} \text{m}$</td>
</tr>
<tr>
<td>$1 \text{ Torr}$</td>
<td>$133.32 \text{ Nm}^{-2}$</td>
</tr>
<tr>
<td>$T$</td>
<td>$363.16 \text{ K}$</td>
</tr>
<tr>
<td>$\sigma_{\text{Cs-Cs}}$</td>
<td>$1.247 \times 10^{-18} \text{m}^2$</td>
</tr>
<tr>
<td>$v_{\text{Cs}}$</td>
<td>$150.185 \text{ m/s}$</td>
</tr>
<tr>
<td>$L$ (length of Cs-cell)</td>
<td>$5.08 \text{ cm}$</td>
</tr>
</tbody>
</table>

4.2.3 Laser Power Effect

We took our polarization data at various temperatures of the cesium cell. At each temperature we set the power by dye laser 1 at three or four different values while keeping the power of dye laser 2 constant and took polarization data. For same set of laser 1 powers, we attenuated the power of laser 2 some other value and took the data. The process resulted with a set of polarization data that could be plotted to analyze the power dependence of dye laser with the polarization degree of the emission spectra. The Fig. 4.1 and Fig. 4.2 show the variation of linear polarization with the laser powers. Within the statistical error, there was no significant power dependence on the measured polarization of the transition.

Figure 4.1: Linear polarization versus power of laser 1 at a constant power of dye laser 2.

4.2.4 Temperature Effect

In order to check to the density dependence of the polarization, we took data at various temperature of the cesium cell. Fig. 4.3 shows a plot of the polarization
Figure 4.2: Linear polarization versus power of laser 2 at a constant power of dye laser 1.

versus temperature. Within the statistical error, the measured linear polarization was independent with the temperature of cesium cell.

Figure 4.3: Linear polarization degree at various temperature of cesium cell.

4.3 Alignment Calculation

We calculated the average polarization percentage from data we took at various temperature and powers of dye lasers for pure cesium atoms. The variation of temperature and power of lasers has no significant effect on our polarization. The polarization for our experiment was found to be 60.52% within the statistical error of 13%. By using the Eq. 2.16 we determined the value of alignment in $6p^2P_{3/2}$ level to be -0.8 with error of 21.7%. The experimentally determined value for
electric quadrupole moment in $6p^2P_{3/2}$ level was quite equal to the theoretical value of -0.8 with 60% polarization.

### 4.4 Depolarization with Krypton Gas

After taking data for pure cesium atoms, we introduced krypton gas into the cesium cell and measured the linear polarization at various buffer gas pressure. Due to the collision of excited cesium atoms with the buffer gas, there was a strong collisional depolarization effect on the polarization of the spectrum. The pressure and polarization data with the statistical errors for our transition is listed in Table 4.3. The Fig. 4.4 shows the dependence of the measured polarization degree on the buffer gas pressure.

![Figure 4.4: Depolarization effect on linear polarization in presence of buffer gas.](image)

### 4.5 Extraction of Depolarization Cross-section

We measured the linear polarization at different pressure of krypton gas ranging from 0 to 100 Torr. Fig. 4.4 shows the pressure dependence of polarization at 90°F. The atomic transition from $6s^2S_{1/2} \rightarrow 10s^2S_{1/2}$ can be written in terms of rate equation and alignment. The collisional depolarization takes place in $6p^2P_{3/2}$ state. So it is sufficient to consider this state for the calculation of depolarization cross-section. The total time dependent population density in any magnetic sublevel can be written as,

$$N(t) = \frac{2\Gamma_p}{\gamma}(1 - e^{-\gamma t})$$  \hspace{1cm} (4.11)

where $\gamma$ and $\Gamma_p$ are the radiative and pump pulse decay rates.

and the alignment can be written as,

$$\langle A_0(t) \rangle = -\frac{8}{5} \frac{\Gamma_p}{\gamma_T}(1 - e^{-\gamma_T t})$$  \hspace{1cm} (4.12)
Table 4.3: Depolarization with Krypton buffer gas

<table>
<thead>
<tr>
<th>Krypton Pressure (Torr)</th>
<th>Polarization (%)</th>
<th>Error (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>60.52</td>
<td>13</td>
</tr>
<tr>
<td>13</td>
<td>29.21</td>
<td>4.4</td>
</tr>
<tr>
<td>40</td>
<td>16.42</td>
<td>20</td>
</tr>
<tr>
<td>71</td>
<td>12.94</td>
<td>43</td>
</tr>
<tr>
<td>81</td>
<td>7.65</td>
<td>26</td>
</tr>
<tr>
<td>92</td>
<td>2</td>
<td>22</td>
</tr>
</tbody>
</table>

The population densities for the magnetic sublevels corresponding to $m = 1/2$ and $m = 3/2$ can be written as,

$$\hat{N}_{1/2}(t) = \frac{N(t) - \frac{10}{\hbar} \langle A_0(t) \rangle g^{(2)}}{4}. \quad (4.13)$$

and

$$\hat{N}_{3/2}(t) = \frac{N(t) + \frac{10}{\hbar} \langle A_0(t) \rangle g^{(2)}}{4}. \quad (4.14)$$

Substituting the values of $N(t)$ and $\langle A_0(t) \rangle$ from Eq. 4.11 and Eq. 4.12 into Eq. 4.13 and Eq. 4.14, the population densities in $m = 1/2$ and $m = + 3/2$ levels will be,

$$\hat{N}_{1/2}(t) = \frac{\Gamma_P}{2} \left[ \frac{1}{\gamma} (1 - e^{-\gamma t}) + \frac{1}{\gamma T} (1 - e^{-\gamma T t})g^{(2)} \right]. \quad (4.15)$$

$$\hat{N}_{3/2}(t) = \frac{\Gamma_P}{2} \left[ \frac{1}{\gamma} (1 - e^{-\gamma t}) - \frac{1}{\gamma T} (1 - e^{-\gamma T t})g^{(2)} \right]. \quad (4.16)$$

For the sake of simplicity, let $\frac{\Gamma_P}{\gamma} (1 - e^{-\gamma t}) = P_1$ and $\frac{\Gamma_P}{\gamma T} (1 - e^{-\gamma T t})g^{(2)} = P_2$. Eqs. 4.15 and 4.16 change into

$$\hat{N}_{1/2}(t) = \frac{1}{2} (P_1 + P_2) \quad (4.17)$$

$$\hat{N}_{3/2}(t) = \frac{1}{2} (P_1 - P_2) \quad (4.18)$$

The parallel signal from the $m = \frac{1}{2}$ quantum state for $\Delta m = 0$ transition is given by,

$$S_{||} = \frac{1}{3} \int_0^T \hat{N}_{1/2}(t) \, dt \quad (4.19)$$

Similarly the perpendicular signal from the $m = \frac{1}{2}$ and $m = \frac{3}{2}$ quantum states for $\Delta m = \pm 1$ transitions can be written as,

$$S_{\perp} = \frac{1}{4} \int_0^T \hat{N}_{3/2}(t) \, dt + \frac{1}{12} \int_0^T \hat{N}_{1/2}(t) \, dt \quad (4.20)$$
Substituting the values of $N_{1/2}$ and $N_{3/2}$ from Eq. 4.17 and Eq. 4.18 into the Eq. 4.19 and Eq. 4.20, we get

$$S_{\parallel} = \frac{1}{6} \int_{0}^{T} P_1 \, dt + \frac{1}{6} \int_{0}^{T} P_2 \, dt$$  \hspace{1cm} (4.21)

$$S_{\perp} = \frac{1}{6} \int_{0}^{T} P_1 \, dt - \frac{1}{12} \int_{0}^{T} P_2 \, dt$$  \hspace{1cm} (4.22)

The linear polarization, as defined earlier, can be found with the following equation

$$P_L = \frac{S_{\parallel} - S_{\perp}}{S_{\parallel} + S_{\perp}}$$  \hspace{1cm} (4.23)

where

$$S_{\parallel} - S_{\perp} = \frac{1}{4} \int_{0}^{T} P_2 \, dt$$  \hspace{1cm} (4.24)

and

$$S_{\parallel} + S_{\perp} = \frac{1}{3} \int_{0}^{T} P_1 \, dt + \frac{1}{12} \int_{0}^{T} P_2 \, dt$$  \hspace{1cm} (4.25)

By evaluating the integrations above and simplifying the algebra we can write the linear polarization as

$$P_L = \frac{3}{4} \frac{Z}{4 + Z}$$  \hspace{1cm} (4.26)

where

$$Z = \frac{g^{(2)} \gamma}{\gamma T} \left[ \frac{1 - \frac{1}{\gamma T} (1 - e^{-\gamma T})}{1 - \frac{1}{\gamma T} (1 - e^{-\gamma T})} \right]$$  \hspace{1cm} (4.27)

The radiative decay rate $\gamma$ of the $6p^2P_{3/2}$ intermediate state is the reciprocal of the life time of the state and can be calculated as,

$$\gamma = \frac{1}{\tau_{6p_{3/2}}}$$  \hspace{1cm} (4.28)

$T$ is the temporal pulse width of the laser and $g^{(2)}$ is the hyperfine depolarization coefficient. The quantity $\gamma_T$ is given by

$$\gamma_T = \gamma + \Gamma$$  \hspace{1cm} (4.29)

where

$$\Gamma = \frac{\rho_{Kr} \cdot \sigma_d \nu_{Kr} - C_s}{P}$$  \hspace{1cm} (4.30)

$$= \frac{\rho_{Kr} \cdot \sigma_d \nu_{Kr} - C_s}{k_B T_c}$$

where $\sigma_d$ is the disalignment cross section, $P$ is the buffer gas pressure, $k_B T_c$ is the thermal energy constant with $k_B$ the Boltzmann’s constant and $T_c$ the absolute
Table 4.4: Important physical quantities used to extract the depolarization cross-section.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$m_{Cs}$</td>
<td>$2.223 \times 10^{-25}$ kg</td>
</tr>
<tr>
<td>$m_{Kr}$</td>
<td>$1.401 \times 10^{-25}$ kg</td>
</tr>
<tr>
<td>$\mu_{Kr-Cs}$</td>
<td>$8.594 \times 10^{-26}$ kg</td>
</tr>
<tr>
<td>$\tau_{6P_{3/2}}$</td>
<td>30.67 ns</td>
</tr>
<tr>
<td>$\gamma_{6P_{3/2}}$</td>
<td>$3.26 \times 10^4$ s$^{-1}$</td>
</tr>
<tr>
<td>$\bar{v}_{Kr-Cs}$</td>
<td>385.37 m/s</td>
</tr>
<tr>
<td>$g^{(2)}$</td>
<td>0.219</td>
</tr>
<tr>
<td>$T$(pulse width)</td>
<td>$6 \times 10^{-9}$ s</td>
</tr>
<tr>
<td>$T_c$(temperature of the cell)</td>
<td>363.16 K</td>
</tr>
<tr>
<td>$k_B$(Boltzmann’s constant)</td>
<td>$1.3807 \times 10^{-23}$ JK$^{-1}$</td>
</tr>
<tr>
<td>1 Torr</td>
<td>133.32 Nm$^{-2}$</td>
</tr>
</tbody>
</table>

The temperature of the cesium cell and $\bar{v}_{Kr-Cs}$ is the average velocity of the Cs-Kr atoms which can be found from the equation

$$\bar{v} = \sqrt{\frac{8k_B T_c}{\pi \mu}}$$  \hspace{1cm} (4.31)

where $\mu$ is the reduced mass of the Cs-Kr atoms.

Substituting the values of all known quantities in Eq. 4.27 and the value of $Z$ so obtained in Eq. 4.26, we get a mathematical equation that relates the linear polarization as a function of buffer gas pressure with $\sigma_d$ the only unknown constant parameter to be determined. We extracted the collisional depolarization cross-section by using a weighted non-linear least square fit as shown in Fig.4.4. The calculated value of collisional depolarization cross-section of the $6p^2P_{3/2}^3$ was about 290 $\text{A}^2$. The alignment depolarization cross section we report here in the $J=3/2$ cesium atoms in collision with Kr atoms had not been measured using a two-photon pump-probe technique. We report here, for the first time, alignment depolarization cross section using a double resonance two-photon two-color pump-probe pulse laser technique. Our reported value is in good agreement with the theoretical value reported by [8]. The numerical values for the physical quantities used to extract the depolarization cross-section are listed in Table 4.4. The measured value of the alignment depolarization cross section along with the reported values are shown in Table 4.6.

Table 4.5: Measured values for electrical quadrupole moment and linear polarization degree in $J=3/2$ level.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_0$</td>
<td>-0.8</td>
</tr>
<tr>
<td>$P_L$(th)</td>
<td>60%</td>
</tr>
<tr>
<td>$P_L$(exp)</td>
<td>60.52(±13)%</td>
</tr>
</tbody>
</table>
Table 4.6: Alignment depolarization cross section in the J=3/2 Cs atoms in collision with different noble gas atoms.

<table>
<thead>
<tr>
<th>Buffer gas</th>
<th>Guiry et al. $\sigma_2(A)^2$</th>
<th>Bayram et al. $\sigma_2(A)^2$</th>
<th>Okunevich $\sigma_2(A)^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>86±21</td>
<td>68</td>
<td></td>
</tr>
<tr>
<td>Ne</td>
<td>88±22</td>
<td>123</td>
<td></td>
</tr>
<tr>
<td>Ar</td>
<td>288±72</td>
<td>186±58</td>
<td>238</td>
</tr>
<tr>
<td>Kr</td>
<td></td>
<td>294±47(this work)</td>
<td>273</td>
</tr>
<tr>
<td>Xe</td>
<td>668±167</td>
<td></td>
<td>400</td>
</tr>
</tbody>
</table>

4.6 Conclusion

We conducted an experiment to study the linear polarization spectrum of the atomic cesium in the excited state by employing the pump-probe technique. The ground state cesium atoms were excited to $6p^2P_{3/2}$ state and finally to $10s^2S_{1/2}$ by using a two-photon two-color excitation process. The excited atoms made a transition back to the ground state with the emission of radiation. Since the s-s transition is forbidden, we observed fluorescence resulted from the transition of the atoms from $9p^2P_{1/2}$ state to $6s^2S_{1/2}$ state. We measured the linear polarization for pure cesium atoms. Since the linearly polarized light produces no orientation in the excited state, we measured the experimental value for the alignment which is the only state multipole present in the $6p^2P_{3/2}$ level and was in good agreement with the calculated value. We finally introduced the krypton gas in the cesium cell and measured polarization data at various buffer gas pressures. Our data showed a strong depolarization effect in the presence of buffer gas pressure. The collision between the ground state krypton gas and excited cesium atom resulted with a strong depolarization of the spectrum. We measured the linear polarization at various krypton pressures and finally extracted the depolarization cross section to be 294(47) in the $6p^2P_{3/2}$ state using a rate equation analysis and then did non-linear least square analysis using Statistical Analysis Software (SAS). Our value shows that there is a good agreement with the theoretical value.
Bibliography


Appendix A

Experimental Apparatus

Figure A.1: Boxcar averager/integrator.
Figure A.2: Oscilloscope.

Figure A.3: Data acquisition board.
Figure A.4: Turbo pump.

Figure A.5: Dye circulation system for dye laser 1.
Figure A.6: Krypton Cylinder.

Figure A.7: Dye laser 1 cavity.
Figure A.8: Dye laser 2 cavity.

Figure A.9: Oven and cesium cell.
Figure A.10: Experimental Setup.

Figure A.11: Experimental Setup.
Appendix B

LabVIEW Program

Figure B.1: Nematic LC Digital Interface.
Figure B.2: Micrometer Controller for tuning laser 2

Figure B.3: Get Spectrum program to test for resonance signal.
Figure B.4: Data taking software.

Figure B.5: Polarization calculating software.