

Master's Thesis

Laser frequency stabilization
in a calcium optogalvanic lamp

Louis Pagot

Supervisors: W. Adamczyk, Dr. H. Fernandes Passagem

Group leader: Prof. Dr. J. Home

Trapped Ion Quantum Information Group
Department of Physics, ETH Zürich

March 13, 2023

Abstract

This work investigates the feasibility of locking a laser frequency on the 733 nm transition between the two excited states 4^1P_1 and 4^1D_2 of neutral calcium atoms using a two-color polarization spectroscopy technique. Indeed, since the atoms are initially in the ground state 4^1S_0 , a 423 nm laser is used to pump them into the 4^1P_1 state, then the addition of the 733 nm laser creates a three-level system. The use of polarized beams allows to obtain a signal that can be used to retroact on the laser. The goal is to use this frequency stabilized laser together with a 423 nm laser to realize a two-color magneto-optical trap and thus achieve a sub-Doppler cooling.

To carry out this objective, we first stabilize the 423 nm laser using the polarization spectroscopy technique. We describe the experimental setup and the resulting frequency stability, and we review the theory of the polarization spectroscopy technique.

Moreover, we study the electromagnetically induced transparency and Autler-Townes splitting phenomena in the three-level ladder system $4^1S_0 - 4^1P_1 - 4^1D_2$ of neutral calcium atoms. The fitting of the data we have collected with the theoretical expression requires the prior determination of some parameters such as the optical density and the temperature. Determining the temperature proves to be a tedious task because the frequency range for scanning the 423 nm laser frequency is limited by mode hops.

Furthermore, we stabilize the 733nm laser frequency with a two-color polarization spectroscopy technique. We give a theoretical overview of its working principle as well as the results obtained when performing it. We prove that this technique can be used to achieve a frequency stability of about 1MHz, which makes it conceivable to use this stabilization technique for a two-photon cooling scheme. Finally, we briefly show that by using a similar setup and replacing the 733 nm laser with a 403 nm laser, we are able to form the three-level system $4^1S_0 - 4^1P_1 - 12^1D_2$. Similar to what is demonstrated in this work, this could be exploited to lock the 403 nm laser on the $4^1P_1 \leftrightarrow 12^1D_2$ transition of neutral calcium atoms.

In principle, the two-color polarization spectroscopy technique to stabilize the frequency of a laser on the transition between two excited states could be applied to other three-level system composed of $S - P - D$ Zeeman manifolds, in particular other alkaline earth atoms such as magnesium or strontium.

Acknowledgements

First of all, I would like to thank the Rydberg team members with whom I worked throughout this project. It was a real pleasure working with you as you were always ready to explain to me how I should proceed and to answer my questions.

I am especially grateful to Henry for supervising me all along this project, for teaching me experimental physics in both optics and electronics, and for all the accurate suggestions that helped me during this master's thesis.

I would like to thank to Wojtek for the pertinent questions and clarifying discussions, which pushed me to better understand the subject. Thanks also for giving me a hand for the various programming steps and saving me some headaches.

Thanks to Silvan for his help and the advice on the optics and techniques I used in the lab.

I am also grateful to Manuel who, during his time in the group, helped me several times with electronics.

Finally, thanks to the members of the TIQI group, as it was a pleasure to work in this group, and in particular to Jonathan, who accepted me as a master's student in the group and gave me the opportunity to work on this project.

Contents

1 Introduction	6
1.1 Trapping calcium atoms in a magneto-optical trap	6
1.2 Laser frequency	7
1.2.1 Frequency noise	7
1.2.2 Frequency control	8
1.3 Thesis outline	8
2 Theory	10
2.1 Electric dipole hamiltonian	10
2.2 Atom-light interaction in a two-level system	11
2.2.1 Time independent hamiltonian	12
2.2.2 Density matrix formalism	13
2.2.3 Specific examples of jump operators	14
2.2.4 Steady state of the optical Bloch equations	15
2.2.5 Saturation of the transition	17
2.2.6 Evolution of a monochromatic wave in atomic medium .	18
2.2.7 Doppler broadening	20
2.3 Zeeman manifolds	23
2.3.1 Rabi frequency	23
2.3.2 Spontaneous decay rate	24
2.4 Spectroscopy in a two-level system	25
2.4.1 Saturation spectroscopy	25
2.4.2 Polarization spectroscopy	28
2.5 Absorption drop in a three-level system	31
2.5.1 Three-level ladder system	31
2.5.2 Dark state	33
2.5.3 Absorption coefficient of a three-level ladder system . .	35
2.5.4 Electromagnetically induced transparency	36
2.5.5 Autler-Townes splitting	39
2.5.6 Modelling influence of collisions	41
2.6 Two-color polarization spectroscopy	42
2.6.1 Simple analogy with polarization spectroscopy	42
2.6.2 Output signal	43
2.6.3 Polarized light beams and Zeeman sub-levels	44
2.6.4 Difference of refractive indices	46
3 Experimental Apparatus	49
3.1 Atomic source	49
3.1.1 Optogalvanic lamp	49

3.1.2 Neutral calcium atoms	50
3.2 Ti:Sapphire <i>MSQUARED SolsTiS</i> laser	51
3.3 <i>HighFinesse WS6 - 200 Series</i> wavemeter	52
3.4 423 nm Polarization spectroscopy	53
3.5 Absorption drop in a three-level system	55
3.6 423 nm - 733 nm Two-color polarization spectroscopy	56
4 Results	59
4.1 423 nm Polarization spectroscopy	59
4.1.1 Polarization spectroscopy signal	59
4.1.2 Locking of the 423 nm laser	62
4.2 Absorption drop in a three-level system	65
4.2.1 Data parameters	65
4.2.2 Determining the optical densities	66
4.2.3 Determining the temperatures	69
4.2.4 Fitting the data	72
4.3 423 nm - 733 nm Two-color polarization spectroscopy	75
4.3.1 Two-color polarization spectroscopy signal	75
4.3.2 Locking of the 733 nm laser	77
4.4 Rydberg excitation	81
5 Conclusion	84
Appendix A Simple Case of Lindblad Operator	85
Appendix B Clebsch-Gordan Coefficients	87
Appendix C Polarization Spectroscopy	88
C.1 Output signal	88
C.2 Difference of refractive indices	89
Appendix D Technicalities	92
D.1 Lock in amplifier	92
D.2 Acousto-optic modulator	93
Appendix E Three-Level Ladder System	96
E.1 Limit $\Omega_p \ll \Omega_c, \Gamma_{12}$	96
E.2 Approximated analytical expression	98

1 Introduction

Quantum computing has raised immense hopes among the general public in recent years. Indeed, the possible applications are numerous and would allow significant advances in many fields: chemistry and material science, biology, machine learning [1], among others. These advances would be made possible by the exponential speedup of algorithms and simulations executed on quantum platforms. These promises seem achievable in the medium to long term thanks to the development of various technologies: trapped ions, superconducting circuits, quantum dots, photons and neutral atoms [2] for instance. In particular, Rydberg atoms appear to be prime candidates because of their long coherence time, their strong coupling to light and the use of Rydberg blockade to perform multiple qubit gates or simulate Ising model [3]. In addition, progress has been made in the control of individual neutral atoms using optical tweezers to form large-scale arrays [4] or to move atoms while preserving the entanglement [5].

The goal of the Rydberg team is to experiment with neutral calcium atoms as a possible platform for quantum simulation. Calcium is an alkaline earth element and as such has two valence electrons. Therefore, if one electron is in a Rydberg state, it might be possible to read this state non-destructively by interacting with the core electron. Indeed, the energy shift induced by the Coulomb interaction depends on the state of the Rydberg electron, as already demonstrated for strontium [6].

1.1 Trapping calcium atoms in a magneto-optical trap

To carry out these experiments, the calcium atoms

1. need to be trapped in a vacuum so that they can be manipulated without being expelled by collisions.
2. need to be cooled as low as possible to minimize thermal agitation effects.

To achieve these two objectives, the use of a magneto-optical trap (MOT) was chosen. This last technique was first reported by Steven Chu [7] and earned him the 1997 Nobel Prize in Physics together with Claude Cohen-Tannoudji and William D. Phillips.¹

¹The Nobel Prize in Physics 1997. NobelPrize.org. Nobel Prize Outreach AB 2023. Sun. 5 Feb 2023. <https://www.nobelprize.org/prizes/physics/1997/summary/>

Similar to an optical molasses, a magneto-optical trap is composed of six perpendicular, counter-propagating laser beams that cool the atoms. However, the beams of opposite directions are of opposite circular polarization, which, combined with a magnetic field, allows to trap the atoms. This technique has the great advantage of being very robust to intensity imbalances between the optical beams and to their approximate polarization [8]. The atoms can be cooled to the Doppler temperature

$$T_{Doppler} = \frac{\hbar\Gamma}{2k_B}. \quad (1)$$

For our purposes, the atoms will first be cooled in a 2D magneto-optical trap before being transferred to the 3D MOT, which will limit the pressure and thus the collisions in the latter. Furthermore, in order to cool the atoms to a temperature below the Doppler limit of 0.8 mK for the simple 423 nm transition between the 4^1S_0 and 4^1P_1 states of neutral calcium atoms, a two-color cooling scheme similar to what has been demonstrated for magnesium [9] will be tested. The objective is to take advantage of the lowest decay rate of the 733 nm transition between the levels 4^1P_1 and 4^1D_2 of the calcium atoms to reach a temperature of about 50 μ K. The realization of this two-color cooling scheme would enable to trap neutral calcium in optical tweezers.

1.2 Laser frequency

In order to realize the two-color cooling technique, the frequencies of the two lasers, 423 nm for the transition between the 4^1S_0 and 4^1P_1 states and 733 nm between the 4^1P_1 and 4^1D_2 states, must be stabilized below 1 MHz. Therefore, the frequency noise of the lasers must be damped.

1.2.1 Frequency noise

For lasers we can distinguish two types of frequency noise: technical noise and quantum noise [10].

The technical noise can have various causes, it can be due to variations in pressure, temperature or mechanical vibrations, among others. Let's take the example of an external cavity diode laser. Its longitudinal modes are determined by the length L of its cavity

$$\nu_n = n \frac{c}{L}, \text{ with } n \text{ an integer.} \quad (2)$$

Thus, a variation dL of the length of the cavity leads to a change in frequency

$$d\nu = -\nu \frac{dL}{L}. \quad (3)$$

For a visible frequency of 600 THz and a 10 cm long cavity, a simple 10 nm change will induce a 60 MHz frequency shift. Consequently, the positions of the cavity mirrors must be very well defined and ideally insensitive or corrected in response to environmental variations such as temperature or pressure drift.

Even if we consider that a laser is perfectly insensitive to technical noise, spontaneous decay being intrinsically stochastic and inherent to the lasing process, it will produce a frequency noise called quantum noise. Indeed, even if the complex amplitude of the laser reaches a steady state through the stimulated emission process, the spontaneous decay generates photons in a random way. This creates amplitude and phase noise. As a result, the laser is no longer a perfectly monochromatic source with a precisely defined frequency, but has a finite linewidth [11]. This linewidth affects the interaction of the laser light with the atoms and acts as a dephasing process.

1.2.2 Frequency control

In order to reduce the frequency noise of a laser, we can first use passive means. This consists essentially in decoupling the laser from its environment in order to reduce the effects of mechanical vibrations by damping them or of temperature variations by insulating it for example. These techniques are generally integrated in the design of lasers [12].

However, active means can also be used. This involves implementing feedback loops on one or more physical variables such as temperature or the position of a mirror in a cavity. Nevertheless, in order to stabilize the frequency, it is interesting to use it directly as a control parameter. Some lasers propose to control the frequency by using a reference cavity or a wavemeter [12]. Though, these two techniques can be subject to drift or can be limited by the precision of the wavemeter, it is therefore interesting to use spectroscopy techniques to have an atomic transition as an absolute reference. It is this last option that we propose to implement to stabilize the lasers that we will use for the two-color cooling scheme.

1.3 Thesis outline

The purpose of this master's thesis is to implement the laser frequency stabilization techniques required for the two-photon cooling scheme. To achieve

this goal, feedback loops with spectroscopy techniques in an optogalvanic lamp are implemented. To stabilize the beam at 423 nm corresponding to the transition between the 4^1S_0 and 4^1P_1 states of neutral calcium atoms, polarization spectroscopy is used. The 733 nm beam is locked to the transition between the 4^1P_1 and 4^1D_2 states using a two-color polarization spectroscopy.

In part **2 Theory**, we introduce the theoretical elements justifying and describing the different experiments that we carry out. We will recall the useful derivations for a two-level system in the semi-classical approach, the electromagnetic field being considered as classical and the internal energy levels of the atoms being quantized. This will give us the theoretical tools to describe the three main experiments we have performed: polarization spectroscopy, absorption drop due to Autler-Townes splitting and finally two-color polarization spectroscopy.

In part **3 Experimental Apparatus**, we describe the implementation of the experiments whose theories have been exposed in the previous part. First, we describe the characteristics of our atomic source which is an optogalvanic lamp. Then, we detail the Ti:Sapphire laser that we use at the wavelength of 733 nm and whose frequency we wish to stabilize as well as the wavemeter that we use to measure the frequency of the laser beams. Finally, we describe the polarization spectroscopy, the absorption drop in the three-level system and finally the two-color polarization spectroscopy experiments.

In part **4 Results**, we gather the results of the three experiments that we have first explained and then described. We fit our theoretical expressions obtained in part **2 Theory**, to the data collected in the setups detailed in part **3 Experimental Apparatus**. In addition, we present some results for the excitation in a low Rydberg state 12^1D_2 of neutral calcium atoms.

2 Theory

2.1 - Electric dipole hamiltonian

Since we will be using laser beams in our experiments, we treat them as classical electromagnetic waves, while retaining the quantized description of atoms. This non-fully quantized treatment is the semi-classical viewpoint [11]. In this subsection, we derive the expression of the electric dipole hamiltonian which accounts for the interaction of the atoms with the electromagnetic field.

For an electromagnetic field describe by the couple vector potential and potential

$$\left(\phi(\vec{r}, t), \vec{A}(\vec{r}, t) \right), \quad (4)$$

in the Coulomb gauge ($\vec{\nabla} \cdot \vec{A} = 0$), the general hamiltonian describing an atom in this field [13] is

$$H = \sum_i \frac{\left[\vec{p}_i - q_i \vec{A}(\vec{r}_i, t) \right]^2}{2m_i} + q_i \phi(\vec{r}_i, t) + \underbrace{\sum_{j < i} \frac{q_j q_i}{4\pi\epsilon_0 |\vec{r}_j - \vec{r}_i|}}_{=V_{Coul}(\{q_i, \vec{r}_i\}_i)}. \quad (5)$$

The sum over the indices i concerns both electrons and protons. The first sum describes the behavior of free particles in the electromagnetic field derived from the potentials (4). The second sum describes the Coulomb interactions between electrons and protons. Due to gauge invariance, the electromagnetic field is the same under the gauge transformation

$$\left(\phi'(\vec{r}, t), \vec{A}'(\vec{r}, t) \right) = \left(\phi(\vec{r}, t) - \frac{\partial}{\partial t} \chi(\vec{r}, t), \vec{A}(\vec{r}, t) + \vec{\nabla} \chi(\vec{r}, t) \right), \quad (6)$$

for any scalar function χ . We can therefore use the Göppert-Mayer transformation [14] using the scalar field

$$\chi(\vec{r}, t) = -\vec{r} \cdot \vec{A}(0, t), \quad (7)$$

where we assume the atom to be at the origin. For an electromagnetic wave, for which $\phi = 0$ because there are no charges, the potentials (6) in the Göppert-Mayer gauge are

$$\left(\phi'(\vec{r}, t), \vec{A}'(\vec{r}, t) \right) = \left(\vec{r} \cdot \partial_t \vec{A}(0, t), \vec{A}(\vec{r}, t) - \vec{A}(0, t) \right). \quad (8)$$

Eventually, the long wavelength approximation leads to the expression of the electric dipole hamiltonian. This approximation consists in neglecting the spacial dependence of the fields, as the typical size of the atom is much smaller than the wavelength of the electromagnetic wave, we can consider that the whole atom is subjected to the same amplitude of the electric field. So we obtain

$$\begin{cases} \vec{A}(\vec{r}, t) \approx \vec{A}(0, t) \\ \vec{E}(\vec{r}, t) \approx \vec{E}(0, t) = -\partial_t \vec{A}(0, t) \end{cases} \quad (9)$$

In the Göppert-Mayer gauge and under the long wavelength approximation, the hamiltonian (5) is

$$H = \sum_i \frac{\vec{p}_i^2}{2m_i} + V_{Coul}(\{q_i, \vec{r}_i\}_i) - \vec{d} \cdot \vec{E}(t), \text{ with } \vec{d} = \sum_i q_i \vec{r}_i. \quad (10)$$

$\vec{d}$ is the dipole operator. The hamiltonian (10) can be decomposed into two components. The first is the atomic hamiltonian in the absence of an external electromagnetic field

$$H_0 = \sum_i \frac{\vec{p}_i^2}{2m_i} + V_{Coul}(\{q_i, \vec{r}_i\}_i), \quad (11)$$

whose eigenenergies correspond to the atomic levels. The second is the electric dipole hamiltonian

$$H_{Dip} = -\vec{d} \cdot \vec{E}(t), \quad (12)$$

which describes the interaction between the electromagnetic field and the atom.

2.2 - Atom-light interaction in a two-level system

In this section, we focus on atom-light interaction in a two-level system. This simple case will allow us to introduce the fundamental theoretical tools we will need to understand the phenomena of polarization spectroscopy, absorption drop in a three-level system and two-color polarization spectroscopy. To do so, we rely throughout this section on the lecture notes [11].

The two-level system is composed of the ground state $|1\rangle$ and the excited state $|2\rangle$, the other states are neglected. These two states are eigenstates of the atomic hamiltonian (11) and their eigenenergies are respectively $\hbar\omega_1$ and $\hbar\omega_2$. Consequently, by choosing the energy offset as the eigenenergy of the ground state the atomic hamiltonian is simply

$$H_0 = \hbar\omega_0 |2\rangle\langle 2|, \quad (13)$$

where $\omega_0 = \omega_2 - \omega_1$. We can as well restrict the dipole operator on the Hilbert space formed by the two states. Due to a symmetry argument its diagonal terms vanish and it becomes

$$\vec{d} = \vec{d}_{12}|1\rangle\langle 2| + \vec{d}_{21}|2\rangle\langle 1|, \quad (14)$$

with $\vec{d}_{12} = \vec{d}_{21}^*$. We consider the simple case where the coupling electromagnetic wave is monochromatic and is described by the electric field

$$\vec{E} = E_0 \cos(\omega t + \phi) \vec{\epsilon}, \quad (15)$$

where ω is the angular frequency and $\vec{\epsilon}$ is the polarization vector.² As a consequence, the electric-dipole hamiltonian (12) describing the atom-light interaction is

$$H_{Dip}(t) = -\vec{d}_{12} \cdot \vec{\epsilon} E_0 \cos(\omega t + \phi) |1\rangle\langle 2| + c.c. . \quad (16)$$

2.2.1 Time independent hamiltonian

The hamiltonian (16) is still time dependent which complicates the analysis of the evolution of the system. To get rid of this dependence, we can first switch to the rotating frame defined by the electromagnetic wave by applying the unitary transformation

$$U = \begin{pmatrix} 1 & 0 \\ 0 & e^{i\omega t} \end{pmatrix}. \quad (17)$$

A change of basis embodied by a unitary transformation like (17) modifies a generic hamiltonian H according to the relation

$$H_U = U H U^\dagger + i\hbar \frac{dU}{dt} U^\dagger \quad (18)$$

Then, the total hamiltonian formed by the atomic hamiltonian (13) and of the electric dipole hamiltonian (16) is transformed into

$$H = \begin{pmatrix} 0 & -\vec{d}_{12} \cdot \vec{\epsilon} E_0 \cos(\omega t + \phi) e^{-i\omega t} \\ -\vec{d}_{21} \cdot \vec{\epsilon} E_0 \cos(\omega t + \phi) e^{i\omega t} & \hbar\delta \end{pmatrix}. \quad (19)$$

$\delta = \omega_0 - \omega$ is the detuning between the resonance frequency of the two levels and the frequency of the electromagnetic field. Eventually, as the

²Note that as we are in the long wavelength approximation (9), the spacial dependence of the electric field is neglected. The eventual constant phase resulting of the term $-\vec{k} \cdot \vec{r}$ is absorbed into the global phase ϕ .

cosines are the sums of complex exponentials, we can make the rotating wave approximation by neglecting the rapidly varying non-diagonal terms of the form $e^{\pm i2\omega t}$. The reason is that these terms will create oscillatory effects at angular frequency 2ω which will cancel on average because they are much faster than the other terms governing the hamiltonian such as $|\delta|$ and $|\vec{d}_{12} \cdot \vec{\epsilon} E_0|$. Another justification is to go back to the description of the atom as a classical dipole, so if it is excited by an electromagnetic wave at the angular frequency ω , we consider that the main response is the one at the same frequency and we neglect the non linear response at the twice the frequency. We define the Rabi frequency as

$$\Omega = -\frac{\vec{d}_{12} \cdot \vec{\epsilon} E_0 e^{i\phi}}{\hbar}. \quad (20)$$

For simplicity, we will consider that the polarization vector $\vec{\epsilon}$ and the phase ϕ are chosen so that the Rabi frequency (20) is real. We finally obtain the time independent hamiltonian

$$H = \hbar \begin{pmatrix} 0 & \frac{\Omega}{2} \\ \frac{\Omega}{2} & \delta \end{pmatrix}. \quad (21)$$

2.2.2 Density matrix formalism

If we considered that the atom together with a perfectly monochromatic laser light was an isolated system, then the wavefunction formalism governed by the Schrödinger equation would be sufficient to describe the unitary evolution of the atom. However, this is not the case, the atom interacts with its environment leading to incoherent evolution and this is why we introduce the density matrix formalism.

In this formalism, the positive and unitary trace density matrix operator describing the atom $\rho = \sum_{i,j} \rho_{i,j} |i\rangle\langle j|$, evolves according to the Lindblad master equation

$$\frac{d\rho}{dt} = \frac{1}{i\hbar} [H, \rho] + \mathcal{L}(\rho). \quad (22)$$

The first term on the right hand side of equation (22) describes the unitary evolution of the density matrix and leads to the same unitary evolution as the Schrödinger equation in the wavefunction formalism. The second term is the Lindblad operator and describes the incoherent evolution of the atom. Indeed, due to Stinespring dilation lemma, see [15], there is always a unitary process to describe the global evolution of a system and its environment, however as we measure the system the projection on its Hilbert space leads

to incoherent processes. The Lindblad operator is defined as

$$\mathcal{L}(\rho) = -\frac{1}{2} \sum_k c_k^\dagger c_k \rho + \rho c_k^\dagger c_k - 2c_k \rho c_k^\dagger, \quad (23)$$

where $\{c_k\}_k$ are jump operators [16] describing the different incoherent processes. To get an idea of the justification of the specific form of the Lindblad operator (23), we develop a toy model in **Appendix A Simple Case of Lindblad Operator**.

The jump operators are usually of the form

$$c_{ij} = \sqrt{\Gamma_{ij}} |i\rangle\langle j|. \quad (24)$$

Such an operator (24) represents an incoherent jump from state $|j\rangle$ to a state $|i\rangle$ at a rate Γ_{ij} .

2.2.3 Specific examples of jump operators

Now that we have the general expression for the Lindblad operator (23), let's use specific examples of jump operators.

First, in quantum optics, one of the most important incoherent process we can account for is spontaneous emission. In the case of a two-level system, the decay from the excited state to the ground state is represented through the jump operator

$$c_{1,2} = \sqrt{\Gamma_{12}} |1\rangle\langle 2|, \quad (25)$$

where Γ_{12} is the linewidth, also called spontaneous decay rate, of the transition. The value of the spontaneous decay rate for the transitions of neutral calcium atoms we consider are detailed in Table 1.

Transition	Wavelength	Decay rate
$4^1P_1 \rightarrow 4^1S_0$	423 nm	34.2 MHz
$4^1D_2 \rightarrow 4^1P_1$	733 nm	2.2 MHz

Table 1: Wavelength and spontaneous decay rate of the transitions we consider in neutral calcium. The numbers are taken from [17].

With this jump operator (25), the corresponding Lindblad operator is

$$\mathcal{L}_{sp}(\rho) = -\frac{1}{2} \Gamma_{12} \{2\rho_{22}|2\rangle\langle 2| + \rho_{12}|1\rangle\langle 2| + \rho_{21}|2\rangle\langle 1| - 2\rho_{22}|1\rangle\langle 1|\}. \quad (26)$$

If we consider the Lindblad master equation (22), the Lindblad operator (26) depletes the population of the excited state in favor of the ground state with

the terms proportional to ρ_{22} . Moreover, due to its incoherent nature, it reduces the coherence between the two levels, see the terms proportional to the non-diagonal elements of the density matrix. A last remark is that if we assume that there is no laser light coupling the two levels $H = 0$ and that initially the atom is in the excited state $\rho_{22} = 1$, then the evolution generated by the Lindblad operator (26) alone and the Lindblad master equation (22) is an exponential decay of the excited state population. This is exactly what is predicted by the Weisskopf-Wigner theory of spontaneous emission [11] due to interactions with the electromagnetic field vacuum modes.³

Then, in addition to spontaneous decay, we can look for dephasing effects. We consider two main causes for these incoherent effects. First, the finite linewidth γ_{las} of the laser we discuss in **1.2.1 Frequency noise**. Second, the collisional dephasing effects associated with a rate γ_{coll} , as we work with optogalvanic lamps, see **3.1.1 Optogalvanic lamp**. We care about these dephasing effects as they limit the drop of absorption in the electromagnetically induced transparency regime [18] that we would like to reach. The jump operator for these processes [11], [19] is

$$c_{dph} = \sqrt{2\gamma_{dph}}|1\rangle\langle 1|, \text{ with } \gamma_{dph} = \gamma_{las} \text{ or } \gamma_{coll}. \quad (27)$$

The Lindblad operator corresponding to both the finite linewidth of the laser and the collisions is

$$\mathcal{L}_{dph}(\rho) = -\gamma_{dph} \{ \rho_{12}|1\rangle\langle 2| + \rho_{21}|2\rangle\langle 1| \}, \text{ with } \gamma_{dph} = \gamma_{las} + \gamma_{coll}. \quad (28)$$

Again, looking at the Lindblad master equation (22), the most significant effect of the operator (28) is to decrease the amplitude of the coherent terms of the density matrix. In addition, it broadens the transition linewidth as we will see in the next subsection.

2.2.4 Steady state of the optical Bloch equations

Now that we have the time-independent hamiltonian (21) and the different components of the total Lindblad operator (26) and (28) we can detail the Lindblad master equation (22) for a two-level system with the different terms of the density matrix operator. These equations are called the optical Bloch equations (OBEs).

³Although the Weisskopf-Wigner theory of spontaneous emission uses the electric dipole hamiltonian (12), it takes into account the quantization of the electromagnetic field, which is beyond the scope of the semi-classical viewpoint we have adopted here.

Denoting $\gamma_{12} = \frac{\Gamma_{12}}{2} + \gamma_{coll} + \gamma_{las}$, the four OBEs for the two-level system (22) are

$$\begin{cases} \frac{d\rho_{11}}{dt} &= -i\frac{\Omega}{2}(\rho_{21} - \rho_{12}) + \Gamma_{12}\rho_{22} \\ \frac{d\rho_{22}}{dt} &= i\frac{\Omega}{2}(\rho_{21} - \rho_{12}) - \Gamma_{12}\rho_{22} \\ \frac{d\rho_{12}}{dt} &= -i\frac{\Omega}{2}(\rho_{22} - \rho_{11}) - (\gamma_{12} - i\delta)\rho_{12} \\ \frac{d\rho_{21}}{dt} &= i\frac{\Omega}{2}(\rho_{22} - \rho_{11}) - (\gamma_{12} + i\delta)\rho_{21} \end{cases} \quad (29)$$

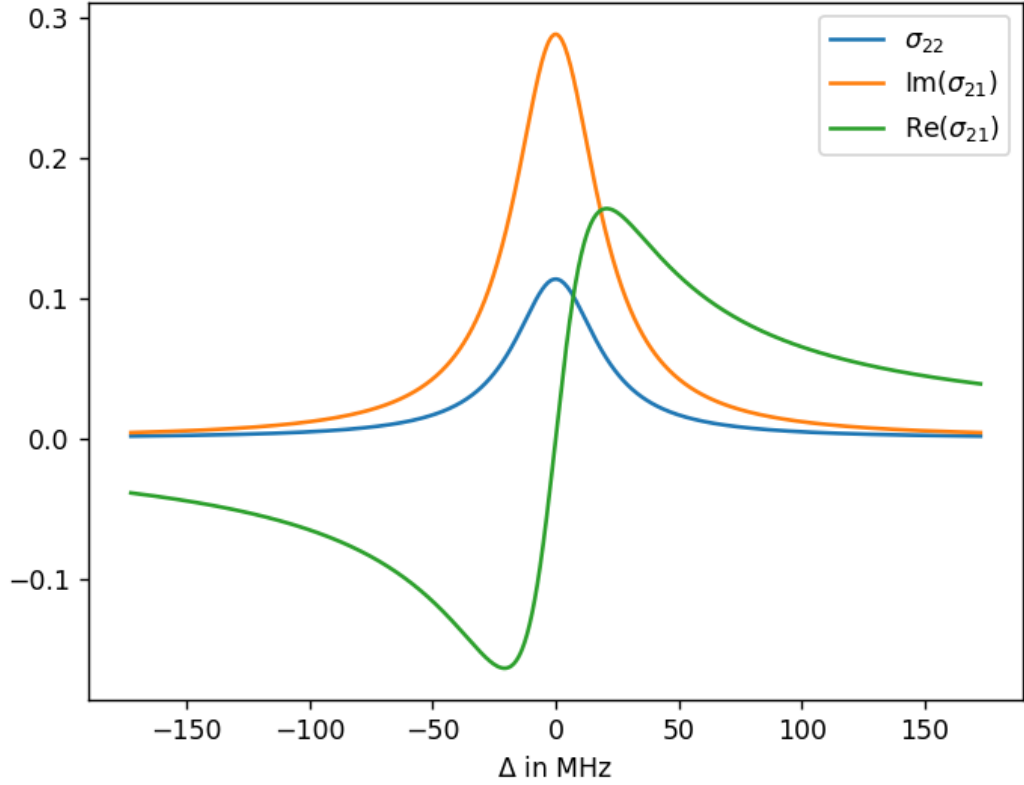


Figure 1: ρ_{22} , $\mathcal{Re}(\rho_{21})$ and $\mathcal{Im}(\rho_{21})$ steady state solutions for the two-level systems (30), as functions of the detuning δ . The parameters are $\Omega = 13.6$ MHz, $\Gamma_{12} = 34.5$ MHz and $\gamma_{coll} + \gamma_{las} = 1$ MHz

Simulations show that the solutions of these equations (29) converge to their steady state in a typical time Γ_{12}^{-1} typically on the order of $1 \mu s$. Because of this fast convergence, we are interested in having a theoretical expression

of the steady state solution of the OBEs (29) by imposing $\partial_t \rho = 0$:

$$\begin{cases} \rho_{12} &= \frac{\Omega}{2} \frac{-\delta + i\gamma_{12}}{\delta^2 + \gamma_{12}^2 + \Omega^2 \frac{\gamma_{12}}{\Gamma_{12}}} \\ \rho_{22} &= \frac{1}{2} \Omega^2 \frac{\gamma_{12}}{\Gamma_{12}} \frac{1}{\delta^2 + \gamma_{12}^2 + \Omega^2 \frac{\gamma_{12}}{\Gamma_{12}}} \\ \rho_{11} &= 1 - \rho_{22} \\ \rho_{21} &= \rho_{12}^* \end{cases} \quad (30)$$

The last two lines are simply the conditions for the density matrix to be a positive and unitary trace operator.

In Figure 1, we can see the steady state solutions of the different terms of the density matrix operator (30) as a function of detuning. The real part of ρ_{21} has a dispersion profile and its imaginary part has an absorption profile. We also see on Figure 1 and with the expressions (30) that the excited state population and the imaginary part of the coherent term are Lorentzian functions centered on zero detuning, which reflects the fact that the coupling is more efficient when the laser is on resonance, and we can define the full width at half maximum (FWHM)

$$\delta_{FWHM} = 2\gamma_s = 2\sqrt{\gamma_{12}^2 + \Omega^2 \frac{\gamma_{12}}{\Gamma_{12}}}. \quad (31)$$

In addition to the plain transition linewidth Γ_{12} , the dephasing processes with rates γ_{las} and γ_{coll} and the use of the laser with a Rabi frequency Ω have broadened the transition linewidth. In the latter case, this phenomenon is known as power broadening [20] and it starts to have a significant effect when the transition is saturated. Thus, the parameter γ_s as defined in (31) is called the saturated linewidth of the transition.

2.2.5 Saturation of the transition

The light intensity is defined as

$$I = \langle ||\vec{E}(\vec{r}, t)||^2 \rangle = \frac{E_0^2}{2}, \quad (32)$$

the last equality being valid for a monochromatic wave which is a good approximation for a single mode laser. With this expression (32), we see that the intensity is proportional to the square of the Rabi frequency (20). Therefore, the power broadening effect we discussed earlier (31) is a function of the beam intensity. To examine the other effects of the saturation of the transition, we introduce the saturation parameter [11]

$$s = \frac{\Omega^2}{\Gamma_{12}\gamma_{12}} \frac{1}{1 + \left(\frac{\delta}{\gamma_{12}}\right)^2} = \frac{s_o}{1 + \left(\frac{\delta}{\gamma_{12}}\right)^2}. \quad (33)$$

s_0 is the maximal value the saturation parameter can reach when the laser is on resonance. It is also defined as the ratio of the light intensity and the saturation intensity

$$s_0 = \frac{I}{I_{sat}}, \text{ with } I_{sat} = \frac{\hbar^2 \Gamma_{12} \gamma_{12}}{2|d_{12}|^2}. \quad (34)$$

The saturated transition linewidth which defines the FWHM (31) is simply

$$\gamma_s = \gamma_{12} \sqrt{1 + s_0}. \quad (35)$$

The steady state solutions of the optical Bloch equations (30) can be expressed in terms of the saturation parameter

$$\begin{cases} \rho_{22} &= \frac{\frac{1}{2} \frac{s}{1+s}}{\frac{\Omega}{2} \frac{-\delta + i\gamma_{12}}{\delta^2 + \gamma_{12}^2} \frac{1}{1+s}} \\ \rho_{12} &= \end{cases} \quad (36)$$

The other coefficients of the density matrix can be deduced from the positivity and unit trace constraints of the density matrix operator. The population in the excited state $|2\rangle$ goes from $\rho_{22}^0 = 0$, when there is no electromagnetic wave, to $\frac{1}{2} \frac{s}{1+s}$, when there is a field which couples the two states via absorption and stimulated emission processes. In the case of a strong saturation: $s \gg 1$ the ratio $\frac{s}{1+s}$ tends towards 1. Thus, the two populations population ρ_{11} and ρ_{22} converge to $\frac{1}{2}$, the population of the ground state $|1\rangle$ being always higher and the one of the excited state $|2\rangle$ being always lower than this value, see (36).

2.2.6 Evolution of a monochromatic wave in atomic medium

Now that we have examined the evolution of the two-level atom under the influence of the electromagnetic field and find its steady state (30), we can look at the evolution of the electromagnetic field itself as it passes through an atomic medium and interacts with the atoms. Classically, with the Lorentz model, see [11], atoms can be described as dipoles so that the atomic medium obtains a polarization, which depends on the applied field and influences in return the propagation of the field. Similarly, in the quantum case, when the light couples the two internal levels of the atoms, their electronic structures evolve over time so that they acquire a non-zero dipole moment. By analogy with the classical case, we can then define the dielectric polarization of the atomic medium and study its effect on the electromagnetic field.

First, since the steady state solution ρ^{st} of the optical Bloch equations (30) is computed in the rotating frame of the electromagnetic wave, we start

by inverting the basis change (17), such that the density matrix operator becomes

$$\rho(t) = \begin{pmatrix} \rho_{11}^{st} & \rho_{12}^{st} e^{i\omega t} \\ \rho_{21}^{st} e^{-i\omega t} & \rho_{22}^{st} \end{pmatrix}. \quad (37)$$

We introduced the superscript st to show the effect of the basis change, but as we do not need it anymore, we remove it. Then, the mean value of the dipole moment is

$$\langle \vec{d} \rangle = \text{Tr} \left(\rho \vec{d} \right) = \rho_{21} \vec{d}_{12} e^{-i\omega t} + c.c. . \quad (38)$$

The dielectric polarization is defined as

$$\vec{P} = \frac{\Delta N}{V} \langle \vec{d} \rangle = \epsilon_0 \chi \vec{E}_0 e^{-i\omega t} + c.c. , \quad (39)$$

where $\frac{\Delta N}{V}$ is the population difference between the ground and the excited states per unit volume, and χ is the linear complex dielectric susceptibility.

Note that in the case of a non-saturating beam, the population difference per unit volume is simply the atomic density because $\rho_{11} - \rho_{22} \approx 1$. For simplicity, we will limit ourselves to this simple case, unless otherwise stated. Indeed, in some cases, such as for polarization spectroscopy, the population difference would be different as the pump beam saturates the transition. In these cases, we will have to multiply the atomic density $\frac{N}{V}$ by the population difference $\rho_{11} - \rho_{22} \neq 1$ [20].

It then follows from the expression of the Rabi frequency (20), the one of the coherent term (36), and the definition of the polarization (39) that the linear complex dielectric susceptibility is

$$\chi = -\frac{N}{V} \frac{|d_{12}|^2}{\hbar \epsilon_0} \frac{\rho_{21}}{\Omega} = \frac{N}{V} \frac{|d_{12}|^2}{\hbar \epsilon_0} \frac{1}{2} \frac{\delta + i\gamma_{12}}{\delta^2 + \gamma_{12}^2 + \Omega^2 \frac{\gamma_{12}}{\Gamma_{12}}} = \chi' + i\chi'' . \quad (40)$$

χ' and χ'' are respectively the real and imaginary part of the linear dielectric susceptibility. Now that we have an expression for the linear complex dielectric susceptibility (40), we can deduce the expression for the refractive index from Maxwell's relation [11]

$$\vec{\Delta} \vec{E} = \frac{1}{c^2} (1 + \chi) \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{n^2}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2}. \quad (41)$$

So that, since the linear dielectric susceptibility is generally small, the refractive index is, to a good approximation,

$$n = \sqrt{1 + \chi} \approx 1 + \frac{\chi}{2} = n' + in'' . \quad (42)$$

Again, we mark the difference between the real and imaginary part with the superscripts ' and ". Therefore, the electric field evolves through the atomic medium as

$$\vec{E}(\vec{r}, t) = \vec{\mathcal{E}} e^{-i(\omega t - n' \frac{\omega}{c} z)} e^{-n'' \frac{\omega}{c} z} + c.c. . \quad (43)$$

The real part of the susceptibility (40), or equivalently of the refractive index (42), defines the dispersion of the wave and the imaginary parts of these quantities define the absorption.⁴

2.2.7 Doppler broadening

For the moment, we have only considered fixed atoms but they can also be in motion and have a non zero velocity $\vec{v}$ as it will be the case in optogalvanic lamps. Then, each atom interacts with the electromagnetic field and undergoes a Doppler shift. In their reference frame, the applied electromagnetic wave has an angular frequency $\omega - \vec{k} \cdot \vec{v}$. Therefore, for the group of atoms of velocity $\vec{v}$, everything happens as derived in the previous subsections, but with a detuning that is now a function of the velocity

$$\delta(\vec{v}) = \omega_0 - \omega + \vec{k} \cdot \vec{v} = \delta + \vec{k} \cdot \vec{v}. \quad (44)$$

Consequently, the electromagnetic field couples the two internal levels of the atoms of this specific group typically if the norm of the velocity dependent detuning (44) is smaller than the transition linewidth, taking into account the power broadening and the dephasing phenomena

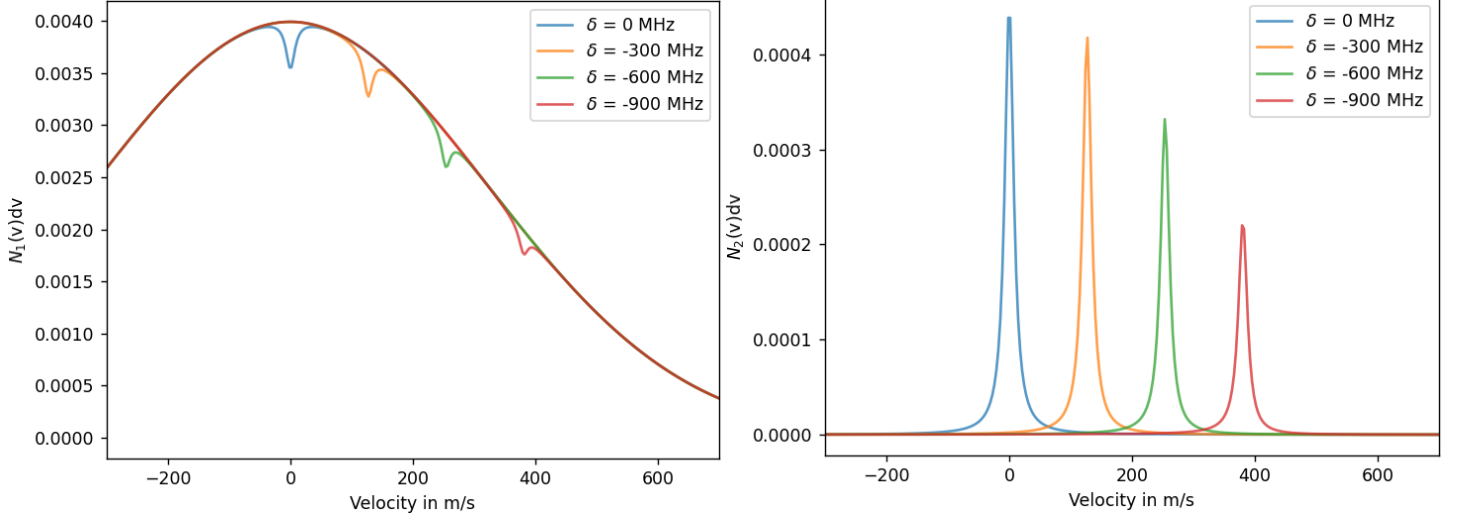
$$\left| \delta + \vec{k} \cdot \vec{v} \right| < \gamma_s. \quad (45)$$

We assume that the field propagates along the z axis and that the probability distribution of the velocity along z is the Maxwell-Boltzmann one

$$w(v_z) = \frac{1}{\sqrt{2\pi}v_{th}} \exp\left(-\frac{v_z^2}{2v_{th}^2}\right) \quad \text{with} \quad v_{th} = \sqrt{\frac{k_b T}{m}}. \quad (46)$$

Then, all the expressions we derived previously and that involve the angular frequency ω of the light beam or equivalently the detuning δ , must be weighted by this probability distribution (46) to take into account the different interactions of the electromagnetic wave with each group of atoms of different velocity.

⁴Note that the amplitude could also be increased, instead of being damped, in the case of a negative imaginary part of the complex refractive index (42). This is the case in population inversion for instance, when before the propagation of the electromagnetic field the population difference is negative $\rho_{11} - \rho_{22} < 0$. This is the operating principle of lasers [11].



(a) Population density of level $|1\rangle$.

(b) Population density of level $|2\rangle$.

Figure 2: Population densities for the two levels (47) and for different detunings of the laser frequency. $\Omega = 13,6$ MHz, $\Gamma_{12} = 34.5$ MHz and $\gamma_{coll} + \gamma_{las} = 1$ MHz are the same parameters as the ones used for Figure 1. The wavelength is $\lambda = 423$ nm and the temperature is $T = 500$ K.

For example, we can define the resulting population densities for the two levels

$$N_i(v_z) dv_z = \rho_{ii}(v_z) w(v_z) dv_z. \quad (47)$$

These quantities (47) describe how many atoms are in the level $|i\rangle$ as a function of their velocity. On Figure 2, we plot the densities for both levels. We see that indeed, the interaction of the atoms with the laser light depletes the population density of the ground state when the condition (45) on the detuning and the Doppler shift is satisfied. This is called a Bennet hole and, when we consider the population density of the excited state, a Bennet peak [21].

Another instance concerns the linear complex dielectric susceptibility which becomes

$$\chi_{Doppler} = \int \chi(\omega - kv_z) w(v_z) dv_z. \quad (48)$$

χ is the previous expression of the linear dielectric susceptibility (40) in the static case where the detuning is replaced by its velocity dependent expression (44). Considering the imaginary part of the dielectric susceptibility (48), which is proportional to the absorption coefficient, it is a convolution of a Lorentzian function and a Gaussian function called a Voigt profile. In the

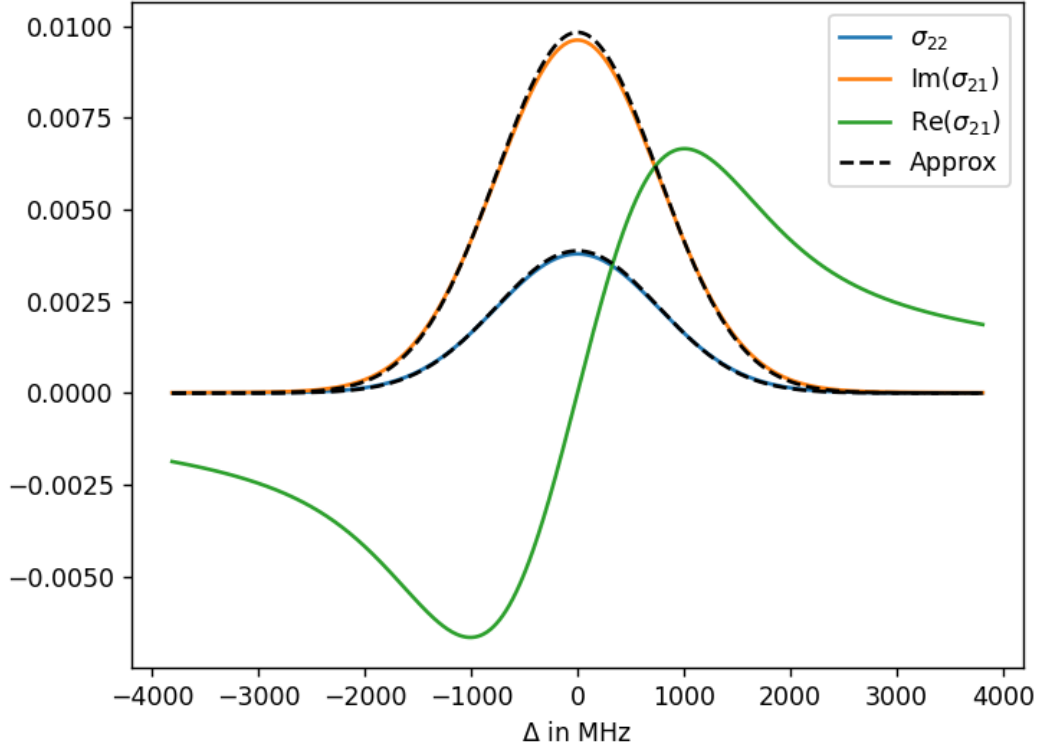


Figure 3: ρ_{22} , $\text{Re}(\rho_{21})$ and $\text{Im}(\rho_{21})$ taking into account Doppler broadening, as functions of the detuning δ . Exact (colored) and the approximated gaussian expressions (black dashed) obtained as in expression (49). $\Omega = 13,6$ MHz, $\Gamma_{12} = 34.5$ MHz and $\gamma_{coll} + \gamma_{las} = 1$ MHz are the same parameters as the ones used for Figure 1. The wavelength is $\lambda = 423$ nm and the temperature is $T = 500$ K.

case of an atomic vapor of hundreds K such that the Gaussian is much larger than the Lorentzian, i.e. $kv_{th} \gg \gamma_s$, we can approximate the Lorentzian as a Dirac function and find an analytical approximation

$$\begin{aligned} \chi''_{Doppler} &= \frac{N}{V} \frac{|d_{12}|^2}{\hbar \epsilon_0} \frac{\pi \gamma_{12}}{2\gamma_s} \int \frac{dv}{\sqrt{2\pi v_{th}^2}} e^{\left(-\frac{v^2}{2v_{th}^2}\right)} \underbrace{\frac{1}{\pi} \frac{\gamma_s}{(\delta + kv)^2 + \gamma_s^2}}_{=\delta_{Dirac}(\omega_0 - \omega + kv)} . \\ &= \frac{N}{V} \frac{|d_{12}|^2}{\hbar \epsilon_0} \frac{\pi \gamma_{12}}{2\gamma_s} \frac{1}{\sqrt{2\pi} kv_{th}} \exp\left(-\frac{(\omega_0 - \omega)^2}{2(kv_{th})^2}\right) \end{aligned} \quad (49)$$

Thus, the absorption coefficient is approximately a Gaussian function of the detuning [21] and its width is the Doppler width kv_{th} . This width is much larger than the natural or saturated linewidth as we can see by comparing the curves on Figure 1 without Doppler broadening and the ones on Figure 3

where Doppler shifts are taken into account, all others parameters being the same. In Figure 3, the imaginary part of the coherent term and the excited state population are plotted for both the exact integration (colored lines) and the approximated one (black dashed lines) as performed in (49). The typical order of the full width at half maximum is $2kv_{th} \approx 1600$ MHz and the amplitudes are lower compare to the curves on Figure 1.

2.3 Zeeman manifolds

As explained in **1 Introduction**, we want to use a two-color polarization spectroscopy technique to stabilize the frequency of a 733 nm laser on the transition between the state 4^1P_1 and the state 4^1D_2 of neutral calcium atom. Hence, by using specific polarization of the lasers we create different coupling strengths between the Zeeman sub-levels of the manifolds. In this section, we study how the Rabi frequency and the spontaneous decay are affected when we consider the the Zeeman sub-levels. The coefficients needed in the case of the Zeeman manifolds $\{4^1S_0, 4^1P_1, 4^1D_2\}$ of neutral calcium atom we consider, are given in **Appendix B Clebsch-Gordan Coefficients**

2.3.1 Rabi frequency

From the expression (20) of the Rabi frequency in the simple case of a two-level system, we can generalize the definition of the Rabi frequency to two states

$$|J_i, m_i\rangle, \text{ and } |J_j, m_j\rangle \quad (50)$$

of the Zeeman manifolds I and J , which is

$$\Omega_{ji} = - \left\langle J_j, m_j \left| \frac{\vec{d} \cdot \vec{E}_0}{\hbar} \right| J_i, m_i \right\rangle. \quad (51)$$

By defining the quantization axis of the atom as the z axis, the dipole operator $\vec{d}$ can be decompose into its three spherical components [22]

$$\vec{d} = d_{-1}\vec{\epsilon}_{-} + d_0\vec{\epsilon}_z + d_{+1}\vec{\epsilon}_{+} \quad \text{with} \quad \vec{\epsilon}_{\pm} = \mp \frac{\vec{e}_x \pm i\vec{e}_y}{\sqrt{2}}. \quad (52)$$

The vectors $\epsilon_{\pm}$ are the polarization vectors of the circular components. According to the Wigner-Eckart theorem, the matrix coefficient of the component s of the dipole operator between the two Zeeman sub-levels (50) is

$$\langle J_j, m_j | d_s | J_i, m_i \rangle = \underbrace{\langle j | d | i \rangle}_{=d_{JI}} \cdot \langle J_j, m_j | J_i, 1; m_i, s \rangle, \quad (53)$$

where d_{JI} is a reduced matrix element and $\langle J_j, m_j | J_i, 1; m_i, s \rangle$ is a Clebsch-Gordan coefficient [22]. d_{JI} depends only on the choice of the Zeeman manifolds I and J and is independent of the sub-levels we consider. For instance, it is the value d_{21} that we introduce in the expression (20) of the Rabi frequency for a two-level system. Therefore, we introduce the notation

$$\Omega_{JI} = -\frac{d_{JI}E_0}{\hbar}. \quad (54)$$

In the experiments, we are interested in two specific cases, when the electromagnetic field is linearly polarized, for simplicity we consider the polarization along the x axis,

$$\vec{E}_0 = E_0 \frac{1}{\sqrt{2}} (\vec{\epsilon}_- - \vec{\epsilon}_+) \quad (55)$$

and when the electromagnetic field is circularly polarized, for simplicity we consider the circular right polarization,

$$\vec{E}_0 = E_0 \vec{\epsilon}_+. \quad (56)$$

In the case of the linearly polarized beam (55), the Rabi frequency between the two Zeeman sub-levels (50) is

$$\Omega_{ji} = \Omega_{JI} \frac{1}{\sqrt{2}} (\langle J_j, m_j | J_i, 1, m_i, -1 \rangle - \langle J_j, m_j | J_i, 1, m_i, +1 \rangle), \quad (57)$$

and for the circular polarized beam, it is

$$\Omega_{ji} = \Omega_{JI} \langle J_j, m_j | J_i, 1, m_i, +1 \rangle. \quad (58)$$

Eventually, to determine the effective Rabi frequencies between the Zeeman sub-levels of the manifolds I and J in the cases of the linearly and circularly polarized beams, (57) and (58), we simply need to calculate the appropriate Clebsch-Gordan coefficients.

2.3.2 Spontaneous decay rate

The spontaneous decay rates are also affected when we consider the coupling between the Zeeman sub-levels. The spontaneous decay rate from a state $|J_j, m_j\rangle$ to a state $|J_i, m_i\rangle$ is Γ_{ij} . All states $\{|J_j, m_j\rangle\}_{-J_j \leq m_j \leq J_j}$ in a Zeeman manifold J have the same total spontaneous decay rate towards the manifold I . This property is mathematically described by the fact that the quantity

$$\Gamma_{IJ} = \sum_{i \in I} \Gamma_{ij}, \quad (59)$$

is independent of the initial state $|J_j, m_j\rangle$ that we consider in the manifold J . Note also that the total decay rate (59) corresponds to the value we use when we do not consider Zeeman sub-levels as in Table 1. The spontaneous decay rate Γ_{ij} from a state $|J_j, m_j\rangle$ to a state $|J_i, m_i\rangle$ is expressed through the expression (59) as [23]

$$\sqrt{\Gamma_{ij}} = \sqrt{\Gamma_{IJ}} \sum_{q=-1}^1 \begin{pmatrix} J_i & 1 & J_j \\ -m_{j_i} & q & m_{j_j} \end{pmatrix} \sqrt{2J_j + 1}. \quad (60)$$

The term in parenthesis in the expression (60) is the Wigner 3-j symbol, it is related to the Clebsch-Gordan coefficient by the relation

$$(-1)^{-j_1+j_2-M} \sqrt{2J+1} \begin{pmatrix} j_1 & j_2 & J \\ m_1 & m_2 & -M \end{pmatrix} = \langle j_1, j_2; m_1, m_2 | J, M \rangle. \quad (61)$$

We prefer to use the Clebsch-Gordan coefficients since we also use them to determine the effective Rabi frequencies between the Zeeman sub-levels (57) and (58).

2.4 Spectroscopy in a two-level system

Generally speaking, spectroscopy is the study of matter through its interactions with the electromagnetic field, as the spectrum of an atom or molecule tells us a lot about its internal structure. As we want to stabilize the frequency of our lasers on the transitions of neutral calcium atoms, it is natural to choose spectroscopic techniques.

Polarization spectroscopy is a spectroscopy technique well suited for a transition between a ground state and an excited state and is not influenced by Doppler broadening, which is why it is called a Doppler free technique [21]. This feature is particularly useful for locking a laser because Doppler broadening would flattens the signal considerably, as can be seen by comparing the real parts of the coherent terms on Figure 1 and 3, and so the feedback loop would be less sensitive.

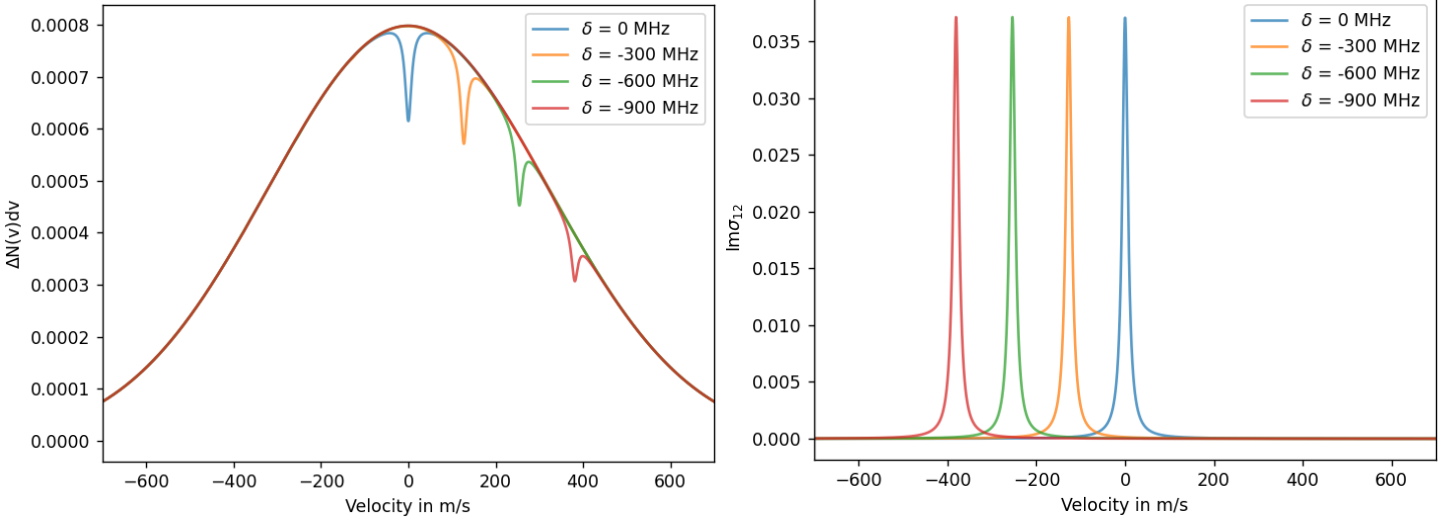
2.4.1 Saturation spectroscopy

To better understand polarization spectroscopy, let us first study the Lamb dip phenomenon, on which the rather similar technique of saturation spectroscopy is based. We will not get into mathematical calculations, as we only want to understand the Doppler-free characteristic of this spectroscopic technique as of polarization spectroscopy.

Assume a saturating pump beam propagating in the $+z$ direction and a weak counter-propagating probe beam, both with the same frequency. According to the condition (45), the pump beam interacts with atoms whose velocity is in the range

$$kv \in I_{pump} = [-\delta - \gamma_s, -\delta + \gamma_s]. \quad (62)$$

This strong pumping beam burns a Bennet hole over this interval (62), as shown in Figure 4 (a), and since the probe beam does not saturate the transition, the population density difference will be defined solely by the pumping beam characteristics.



(a) Difference of population density $\Delta N^{pump} dv_z$ caused by the pump beam.

(b) $\mathcal{I}m(\rho_{21}^{probe})$ due to the probe beam.

Figure 4: Difference of probability densities $\Delta N^{pump} dv_z = (N_1^{pump} - N_2^{pump}) dv_z$ caused by the pump beam and $\mathcal{I}m(\rho_{21}^{probe})$ due to the probe beam for different detunings of the laser frequency. $(\Omega_{pump}, \Omega_{probe}) = (13.6, 1.36)$ MHz, $\Gamma_{12} = 34.5$ MHz and $\gamma_{coll} + \gamma_{las} = 1$ MHz. The wavelength is $\lambda = 423$ nm and the temperature is $T = 500$ K.

Similarly, the probe beam interacts with atoms whose velocity in the range

$$kv \in I_{probe} = [+ \delta - \gamma_{12}, + \delta + \gamma_{12}]. \quad (63)$$

Since the probe beam is the one we are measuring, if we look at the absorption of this beam, then it is defined by the imaginary part of the coherent term ρ_{21} when only this beam is considered. The latter is a Lorentzian on the typical interval (63), see Figure 4 (b).

As we can see in Figure 4, the Bennet hole of the population density difference $\Delta N^{pump} dv_z$ and the imaginary part of the coherent term ρ_{21}^{probe} only overlap when the detuning is approximately zero or equivalently when the beams are interacting with atoms of approximately zero velocity along the z axis. Thus, since the imaginary part of the refractive index is obtained by integrating over the velocities

$$\mathcal{I}m(n) \propto \int \Delta N^{pump}(v_z) dv_z \cdot \mathcal{I}m(\rho_{21}^{probe}), \quad (64)$$

we see by looking at the curves on Figure 4 (a) and (b), that for non zero

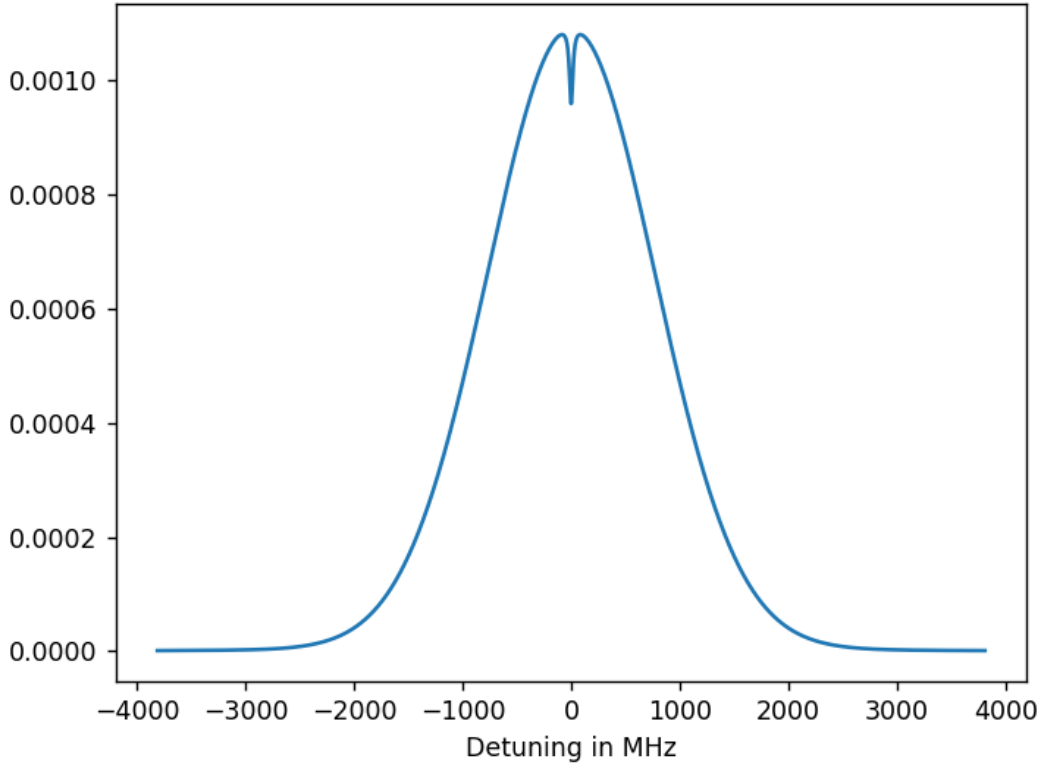


Figure 5: Absorption coefficient up to a proportional factor (64) as a function of the detuning. $(\Omega_{pump}, \Omega_{probe}) = (13.6, 1.36)$ MHz, $\Gamma_{12} = 34.5$ MHz and $\gamma_{coll} + \gamma_{las} = 1$ MHz. The wavelength is $\lambda = 423$ nm and the temperature is $T = 500$ K. These parameters are the same as in Figure 4.

detuning we will obtain a gaussian signal, but that the signal will drop around zero detuning. The exact integration is done numerically in Figure 5. The drop in the absorption coefficient at zero detuning is referred as the Lamb dip. Moreover, the width of the dip is Doppler free and depends, like the

amplitude of the dip, on the degree of saturation of the pump beam. Because of these two properties, this setup can be used as a spectroscopy technique called saturation spectroscopy [21].

2.4.2 Polarization spectroscopy

In this subsection, we directly give the mathematical expressions for the polarization spectroscopy signal and the refractive indices difference, however the derivations are detailed in **Appendix C Polarization Spectroscopy**.

Instead of implementing a simple saturation spectroscopy, we choose to use the polarization spectroscopy technique because it has some advantages such as a better signal-to-noise ratio [21], [24]. The polarization spectroscopy setup shown on Figure 6, is very similar to the one we described in the previous subsection **2.4.1 Saturation spectroscopy**. Indeed, the main idea of using a saturating pump beam to burn a hole in the density population and using a weak counter-propagating beam to probe it and to obtain a Doppler free signal, is exactly the same.

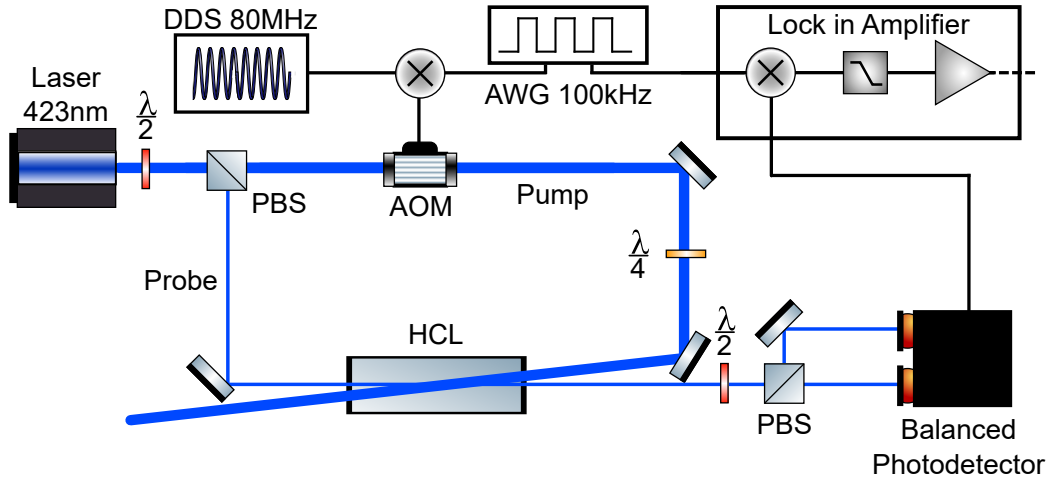


Figure 6: Setup for the polarization spectroscopy⁵. *DDS*: Direct Digital Synthesizer, *AWG*: Arbitrary Waveform Generator, *PBS*: Polarization Beam Splitter, *AOM*: Acousto-Optics Modulator, *HCL*: Hollow Cathode Lamp, $\lambda/2$: half-wave plate, $\lambda/4$: quarter-wave plate.

However, as we can see in Figure 6, we introduce wave plates to specify the polarization of the two beams. Since we take into account the polarization of the beams, we have to consider the Zeeman sub-levels in return. To keep

⁵The figure is made with *ComponentLibrary*. *ComponentLibrary* by Alexander Franzen is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported License.

things simple and stick to our case of interest for neutral calcium atoms, we assume that the transition occurs between a S and a P Zeeman manifolds, as shown in Figure 7, and that the beams propagate along the z axis. We also consider that the pump beam is circular right polarized, as in Figure 7, and that the counter-propagating probe beam that goes to the balanced photo-detector, see Figure 6, is linearly polarized along the x axis.

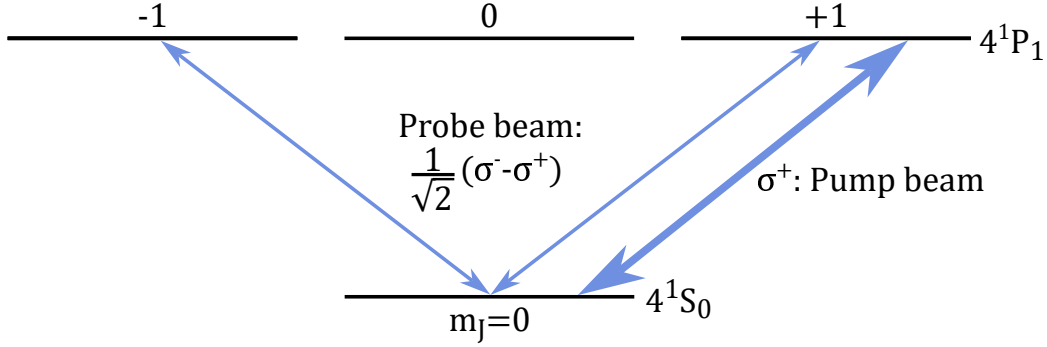


Figure 7: Saturation of the transition $m_0 = 0 \leftrightarrow m_1 = 1$, in the polarization spectroscopy setup for the 423nm transition between the 4^1S_0 and 4^1P_1 states of neutral calcium atoms.

Intuitively, looking at Figure 7, the new degree of freedom we introduce with polarization spectroscopy compared to the Lamb dip phenomenon in a two-level system is the fact that the probe beam now has two components that we can use (sum, subtract) to get a better signal. This is exactly the property that is exploited by the ensemble of the half-wave plate, the polarization beam splitter and the balanced photo-detector placed after the optogalvanic lamp, see Figure 6. The expression of the resulting intensity, derived in **C.1 Output signal**, is

$$\Delta I = \frac{E_0^2}{2} e^{-2\mathcal{I}m(n_{tot} + w_{tot}) \frac{\omega}{c} L} \mathcal{R}e(\Delta n) \frac{\omega}{c} L. \quad (65)$$

$\frac{E_0^2}{2}$ is the initial intensity of the probe beam, L is the length of the optogalvanic lamp and ω is the angular frequency of the laser light. The other quantities are defined as

$$\begin{aligned} n_{tot} &= \frac{n_+ + n_-}{2} & \text{and} & & \Delta n &= n_+ - n_- \\ w_{tot} &= \frac{w_+ + w_-}{2} & \text{and} & & \Delta w &= w_+ - w_- \end{aligned} \quad (66)$$

n_- and n_+ are the refractive indices faced by the left and right circular polarization components of the probe beam as they pass through the atomic

medium in the presence of the pump beam. Likewise, w_- and w_+ are the refractive indices of the optogalvanic lamp windows for the corresponding circularly polarized components of the probe beam.

The useful features for using the signal (65) as an error signal and for locking the laser frequency with it, reside in the real part of the difference of refractive indices. The detailed derivation is given in **C.2 Difference of refractive indices** and is

$$\mathcal{Re}(\Delta n) = -\frac{N}{V} \frac{|d_{12}|^2}{8\hbar\epsilon_0} \frac{\Omega^2\gamma_{12}}{2\Gamma_{12}} \frac{\pi}{\sqrt{2\pi}kv_{th}\gamma_s} e^{-\frac{\delta^2}{2(kv_{th})^2}} \frac{\delta}{\delta^2 + \frac{1}{4}(\gamma_{12} + \gamma_s)^2}. \quad (67)$$

As the gaussian in the expression (67) varies slowly compared to the other part of the expression, we can focus on the latter. This part is interesting because it is very similar to the refractive index expression (42) that we obtain for the two-level system without Doppler broadening, although the full width at half maximum separating the signal extrema is now

$$\delta_{FWHM} = \gamma_{12} + \gamma_s. \quad (68)$$

Since it does not depend on Doppler broadening, polarization spectroscopy is effectively a Doppler free spectroscopy technique. It corresponds to the sum of the FWHMs induced by the probe and the pump beams (31) up to a one half factor appearing when the frequency of both beams is swept at the same time. Compared to the previous Lamb dip case, the gaussian in the polarization spectroscopy signal (67) is simply an envelope function and is not a background signal on which the spectroscopic signal appears as on Figure 5.

The expression (67) is correct, but in order to adapt it to our experiment, we must modify it slightly. Indeed, instead of using a mechanical chopper combined with a lock-in amplifier, we use a chopped acousto-optic modulator (AOM), see **Appendix D Technicalities**. Thus, only the -1 order of the resulting pump beam passes through the optogalvanic lamp. This implies that the detuning of the pump beam have to be replace by $\delta + \delta_{AOM}$, where δ_{AOM} is the frequency of the signal driving the AOM. The final expression is

$$\mathcal{Re}(\Delta n) = -\frac{N}{V} \frac{|d_{12}|^2}{8\hbar\epsilon_0} \frac{\Omega^2\gamma_{12}}{\Gamma_{12}} \frac{\pi e^{-\frac{(\delta+\delta_{AOM})^2}{2(kv_{th})^2}}}{\sqrt{2\pi}kv_{th}\gamma_s} \frac{\delta + \frac{1}{2}\delta_{AOM}}{(\delta + \frac{1}{2}\delta_{AOM})^2 + \frac{1}{4}(\gamma_{12} + \gamma_s)^2}, \quad (69)$$

corresponding approximately to that without the effect of the acousto-optic modulator (67) shifted by $\frac{1}{2}\delta_{AOM}$.⁶

⁶The computation of $\mathcal{Re}(\Delta n)$ with the detuning of the acousto-optic modulator is the same as that detailed in **C.2 Difference of refractive indices**, although using the

2.5 Absorption drop in a three-level system

We detailed in the previous section **2.4 Spectroscopy in a two-level system**, the technique we use to stabilize the 423 nm laser frequency on the transition of neutral calcium atoms between the 4^1S_0 and 4^1P_1 Zeeman manifolds. Since we also want to stabilize the 733 nm laser frequency on the transition between the 4^1P_1 and the 4^1D_2 manifolds, we now have to consider a three-level system. A peculiarity of these systems compared to the two-level systems is that if the two lasers are on resonance, we can observe a drop in absorption. There are two possible causes that we study here: the electromagnetically induced transparency (EIT) and the Autler-Townes splitting (ATS).

2.5.1 Three-level ladder system

To stick to the case of interest, that of neutral calcium atoms, we consider a three-level ladder system, see Figure 8 (a), where the two light beams couple the state $|1\rangle$ and the state $|3\rangle$ by the intervention of another state, state $|2\rangle$, with an intermediate energy.⁷

We derive the time-independent hamiltonian for the three-level ladder system in Figure 8 (a). Since the steps are the same as in section **2.2 Atom-light interaction in a two-level system**, we will not go into the details of the derivation. The time-dependent hamiltonian of the system is the sum of the atomic hamiltonian and the electric-dipole hamiltonian describing the couplings induced by the two electromagnetic fields. Analogously to the expressions (13) and (16), and assuming that the polarization vector and the phase of the two laser beams are correctly chosen, we obtain

$$H(t) = \begin{pmatrix} 0 & -d_{12}E_p(t) & 0 \\ -d_{12}E_p(t) & \hbar\omega_{12} & -d_{23}E_c(t) \\ 0 & -d_{23}E_c(t) & \hbar\omega_{13} \end{pmatrix}. \quad (70)$$

Then, we switch to the rotating frame of the electromagnetic waves by applying the unitary transformation (18) represented by the matrix

$$U = \begin{pmatrix} 1 & 0 & 0 \\ 0 & e^{i\omega_p t} & 0 \\ 0 & 0 & e^{i(\omega_p + \omega_c)t} \end{pmatrix}. \quad (71)$$

residue theorem with the pole $-\delta - \delta_{AOM} + i\gamma_s$.

⁷There are also Λ systems, where the coupling state $|2\rangle$ has an energy level higher than the two other levels. In this case, the physics and mathematics are quite similar [25].

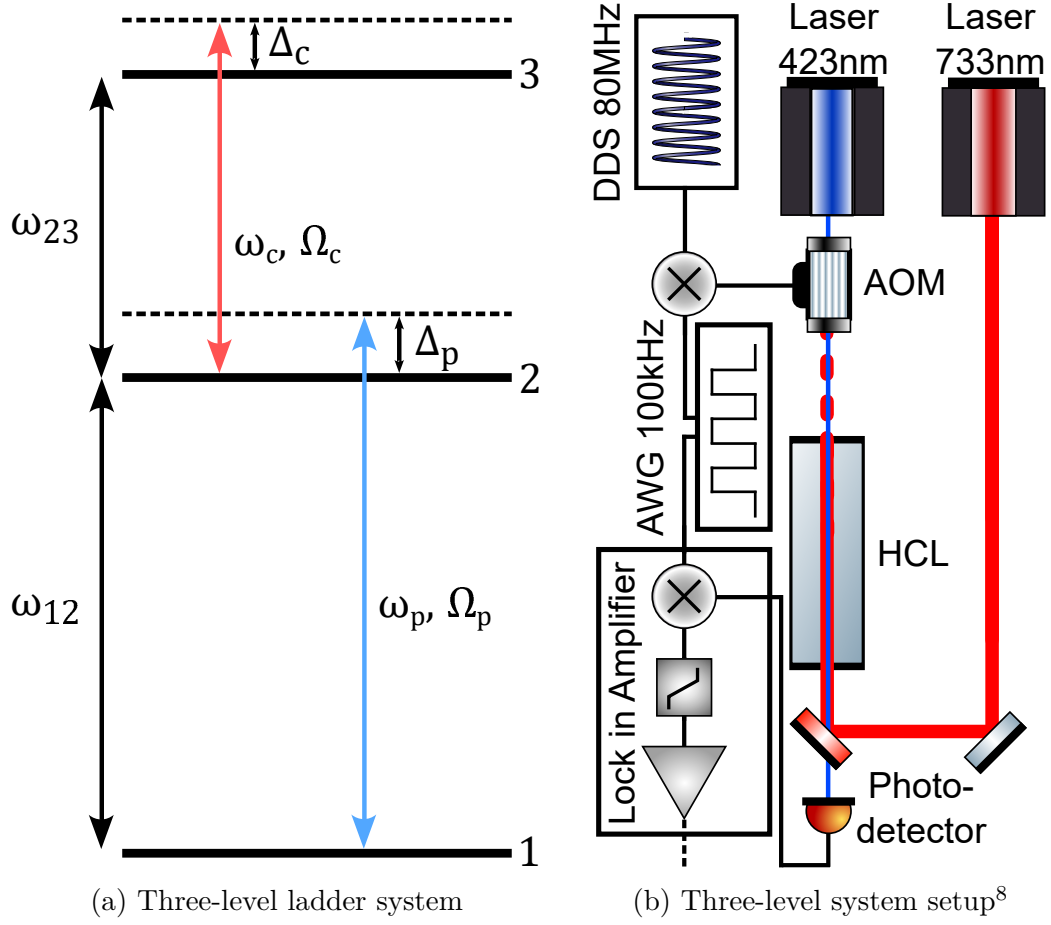


Figure 8: Systems with control and probe fields at frequencies ω_c and ω_p detuned such that $\Delta_c = \omega_c - \omega_{23}$ and $\Delta_p = \omega_p - \omega_{12}$. DDS: Direct Digital Synthesizer, AOM: Acousto-Optics Modulator, HCL: Hollow Cathode Lamp, AWG: Arbitrary Waveform Generator.

Finally, we make the rotating wave approximation and we define the real Rabi frequencies,

$$\Omega_p = -\frac{d_{12}E_{p,0}}{\hbar} \text{ and } \Omega_c = -\frac{d_{23}E_{c,0}}{\hbar}. \quad (72)$$

The subscript p (c) usually stands for *probe* (*control*) and E_{i0} is the $i = p$ or c electric field amplitude. We also define the detunings of the lasers

$$\Delta_p = \omega_{12} - \omega_p \text{ and } \Delta_c = \omega_{23} - \omega_c. \quad (73)$$

⁸The figure is made with *ComponentLibrary*. ComponentLibrary by Alexander Franzen is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported License.

The time independent hamiltonian is

$$H = \hbar \begin{pmatrix} 0 & \frac{\Omega_p}{2} & 0 \\ \frac{\Omega_p}{2} & \Delta_p & \frac{\Omega_c}{2} \\ 0 & \frac{\Omega_c}{2} & \Delta_c + \Delta_p \end{pmatrix}. \quad (74)$$

Furthermore, we take into account incoherent processes, spontaneous decay processes (26) and dephasing processes (28) due to collisions and linewidths of the lasers [19], represented by dephasing rates γ_p and γ_c . The sum of the Lindblad operators is

$$\mathcal{L}(\rho) = \begin{pmatrix} \Gamma_{12}\rho_{22} + \Gamma_{13}\rho_{33} & -\gamma_{12}\rho_{12} & -\gamma_{13}\rho_{13} \\ -\gamma_{12}\rho_{21} & \Gamma_{23}\rho_{33} - \Gamma_{12}\rho_{22} & -\gamma_{23}\rho_{23} \\ -\gamma_{13}\rho_{31} & -\gamma_{23}\rho_{32} & -(\Gamma_{13} + \Gamma_{23})\rho_{33} \end{pmatrix}, \quad (75)$$

where we introduce the quantities

$$\gamma_{12} = \frac{\Gamma_{12}}{2} + \gamma_p + \gamma_c, \quad \gamma_{23} = \frac{\Gamma_{12} + \Gamma_{23} + \Gamma_{13}}{2} + \gamma_c, \quad \gamma_{13} = \frac{\Gamma_{23} + \Gamma_{13}}{2} + \gamma_p. \quad (76)$$

The steady state solutions of the Lindblad master equation (22) for the three-level ladder system are detailed in **Appendix E Three-Level Ladder System**.

2.5.2 Dark state

Before using the density matrix formalism, we describe the dark state phenomenon in the more simple wavefunction formalism in order to have a physical intuition of the electromagnetically induced transparency.

When the frequencies of the two lasers match the two-photons resonance condition

$$\Delta_p + \Delta_c = 0, \text{ or equivalently } \omega_p + \omega_c = \omega_{12} + \omega_{23} = \omega_{13} \quad (77)$$

the state

$$|\psi\rangle = \frac{1}{\sqrt{\Omega_p^2 + \Omega_c^2}} \begin{pmatrix} -\Omega_c \\ 0 \\ \Omega_p \end{pmatrix}, \quad (78)$$

is an eigenstate of the hamiltonian (74) with a corresponding eigenenergy $E = 0$. It is a dark state.

The fact that this eigenenergy is equal to zero is different from the choice of an energy reference as we have done with for the atomic hamiltonian (13) and (70). Because of the change of basis (71), the hamiltonian (74) describes the interaction of the atom and the light beams in the latter's reference frame.

Thus, this zero energy means that the state (78) does not interact with light and this is why it is called a dark state. Mathematically, if we reverse the basis change (71), using the two-photons resonance condition (77), the time evolution of the dark state is

$$|\psi(t)\rangle = \frac{U^\dagger}{\sqrt{\Omega_p^2 + \Omega_c^2}} \begin{pmatrix} -\Omega_c \\ 0 \\ \Omega_p \end{pmatrix} = \frac{-1}{\sqrt{\Omega_p^2 + \Omega_c^2}} (\Omega_c|1\rangle - \Omega_p e^{-i\omega_{13}t}|3\rangle). \quad (79)$$

This time evolution (79), with the fact that the amplitudes for the states $|1\rangle$ and $|3\rangle$ are independent of time, corresponds to the evolution under the Schrödinger equation only considering the atomic hamiltonian. As a reminder and to be even more convinced of the non-interacting behaviour of the dark state, we can consider the same electric-dipole hamiltonian in the case of a two-level system (21). It has eigenenergies $\frac{\Delta}{2} \pm \frac{1}{2}\sqrt{\Delta^2 + \Omega^2}$ and leads to Rabi oscillations. Whatever the initial superposition of the eigenstates of the atomic hamiltonian, the amplitudes of the states $|1\rangle$ and $|2\rangle$ depend on time as the atom interacts with the electromagnetic field.

Note that in the case where the control beam is much stronger than the probe beam, $|\Omega_c| \gg |\Omega_p|$, the dark state (78) is very close to the ground state $|1\rangle$. As the atoms in the dark state do not interact with light, they form a transparent medium, that is why this phenomenon is also called electromagnetically induced transparency because the application of a second electromagnetic wave induces the formation of a transparent medium.

To get a better intuition on how the dark state is formed and why it is a purely quantum state, let us consider the case where both detunings are zero $\Delta_p = \Delta_c = 0$ and take $\hbar = 1$. According to expression (74), the hamiltonian can be decomposed as

$$H = \begin{pmatrix} 0 & \frac{\Omega_p}{2} & 0 \\ \frac{\Omega_p}{2} & 0 & \frac{\Omega_c}{2} \\ 0 & \frac{\Omega_c}{2} & 0 \end{pmatrix} = \underbrace{\begin{pmatrix} 0 & \frac{\Omega_p}{2} & 0 \\ \frac{\Omega_p}{2} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}}_{=H_p} + \underbrace{\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & \frac{\Omega_c}{2} \\ 0 & \frac{\Omega_c}{2} & 0 \end{pmatrix}}_{=H_c}. \quad (80)$$

The hamiltonians H_p and H_c (80) correspond to rotations on the sub-spaces $\{|1\rangle, |2\rangle\}$ and $\{|2\rangle, |3\rangle\}$. To look at the evolution of a quantum state we cannot treat them separately because the intersection of their sub-spaces is non trivial. However, at short time scale dt , small compared to the characteristic time of the two rotations $\tau_i = \Omega_i^{-1}$, the propagator can be developed with the Lie-Trotter formula

$$U(dt) = e^{-i(H_p+H_c)dt} = e^{-iH_pdt}e^{-iH_cdt} + \mathcal{O}(dt^2[H_p, H_c]). \quad (81)$$

Therefore, at this small time scale, the evolution operator (81) act on states $|1\rangle$ and $|3\rangle$ as if the two distinct rotations generated by H_p and H_c were applied

$$\begin{cases} |1\rangle & \rightarrow |1\rangle + i\frac{\Omega_p}{2}dt|2\rangle + \mathcal{O}(dt^2[H_p, H_c]) \\ |3\rangle & \rightarrow |3\rangle + i\frac{\Omega_c}{2}dt|2\rangle + \mathcal{O}(dt^2[H_p, H_c]) \end{cases} \quad (82)$$

From the evolution (82), it is clear that the superposition state (78) formed from the two states $|1\rangle$ and $|3\rangle$ has a zero first time derivative. The coefficients in front of $|1\rangle$ and $|3\rangle$ are chosen in such a way that the small amount of atoms excited by stimulated absorption from $|1\rangle$ to $|2\rangle$ and de-excited by stimulated emission from $|3\rangle$ to $|2\rangle$ have the same amplitude but an opposite phase resulting in a destructive interference process. This is why dark states are purely quantum states for atoms. Such a reasoning can be generalized also in the case where $\Delta_p \neq 0$, the two-photon resonance condition (77) being still met.

Note that dark state can only be formed when there are strictly more than two levels. Indeed, in two-level systems, the hamiltonian can be decomposed with Pauli matrices $\{\sigma_i\}_i$. The propagator of such a hamiltonian is a rotation on the Bloch sphere and sends two orthogonal states on another orthogonal basis. Therefore, it prohibits any possibility of destructive phase interference. This is why we can observe EIT in systems with three levels or more [25].

2.5.3 Absorption coefficient of a three-level ladder system

Assuming that we know the steady state expressions of the coefficient of the density matrix operators, we can study the evolution of the laser beams in the atomic medium in a similar way as for the two-level system. By performing calculation similar to those detailed in **2.2.6 Evolution of a monochromatic wave in atomic medium**, we can determine the mean dipole moment

$$\langle \vec{d} \rangle = \rho_{21}\vec{d}_{12}e^{-i\omega_p t} + \rho_{32}\vec{d}_{23}e^{-i\omega_c t} + c.c., \quad (83)$$

and the dielectric polarization

$$\vec{P} = \frac{\Delta N}{V} \langle \vec{d} \rangle = \epsilon_0 \left(\chi_p \vec{E}_{p,0} e^{-i\omega_p t} + \chi_c \vec{E}_{c,0} e^{-i\omega_c t} + c.c. \right) \quad (84)$$

Since we are interested in the evolution of the probe beam through the atomic vapor, and due to the linearity of the Maxwell's equation (41), we only keep the terms of the dielectric polarization oscillating at the angular frequency ω_p . We end up with the same expression for the refractive index as in the

two-level case (42)

$$n = 1 - \frac{1}{2} \frac{N}{V} \frac{d_{12}^2}{\hbar \epsilon_0} \frac{\rho_{21}}{\Omega_p}. \quad (85)$$

Though, the difference lies in the steady state expression for the coherent term ρ_{21} . We have seen that the imaginary part of the refractive index define the absorption coefficient of the electromagnetic field (43). Consequently, we focus on the expression of the coherent term ρ_{21} and its imaginary part.

2.5.4 Electromagnetically induced transparency

Now that we have studied the dark state phenomenon in the wavefunction formalism, we move to a more realistic approach through the density matrix formalism, as developed in the article [25]. Here, we directly introduce the expression of the coherent term ρ_{12} , which determines the absorption of the probe beam (85), in the case where the control beam is much stronger than the probe beam

$$\Omega_p \ll \Omega_c. \quad (86)$$

The coherent term between state $|1\rangle$ and $|2\rangle$ is then

$$\rho_{12} = i \frac{\Omega_p}{2} \frac{\delta_{13}}{\delta_{13}\delta_{12} + \frac{\Omega_c^2}{4}} = i \frac{\Omega_p}{2} \frac{1}{\delta_{12}} \left(1 - \frac{\Omega_c^2}{4} \frac{1}{\delta_{13}\delta_{12} + \frac{\Omega_c^2}{4}} \right) \quad (87)$$

with $\delta_{12} = \gamma_{12} - i\Delta_p$ and $\delta_{13} = \gamma_{13} - i(\Delta_p + \Delta_c)$. The derivation of this expression (87) and of a more general and more complex expression of the imaginary part of ρ_{21} , as well as the comparison of the expression (87) with the exact solution for different values of the ratio $\frac{\Omega_c}{\Omega_p}$, keeping all other parameters fixed, are done in **Appendix E Three-Level Ladder System**.

Suppose we are in the situation where the control laser is tuned to the resonance of the transition between the state $|2\rangle$ and the state $|3\rangle$, so that $\Delta_c = 0$, and that the frequency of the applied probe beam is swept around the resonance frequency ω_{12} . We will compare the absorption in this case to that of the simple two-level system (30) with a weak probe beam $\Omega_p \ll \gamma_{12}$, such that the coherent term is

$$\rho_{12}^{2lvl} = \frac{\Omega_p}{2} \frac{-\Delta_p + i\gamma_{12}}{\Delta_p^2 + \gamma_{12}^2} = i \frac{\Omega_p}{2} \frac{1}{\delta_{12}}. \quad (88)$$

Thus, in the three-level case (87), we have an additional function that multiplies the two-level expression (88). Let's see the modifications it induces.

In the first case, $\Delta_p \gg \gamma_{12}$ as the right term in the parenthesis of the expression (87) has a Δ_p^{-2} dependence, we can neglect it and we end up with

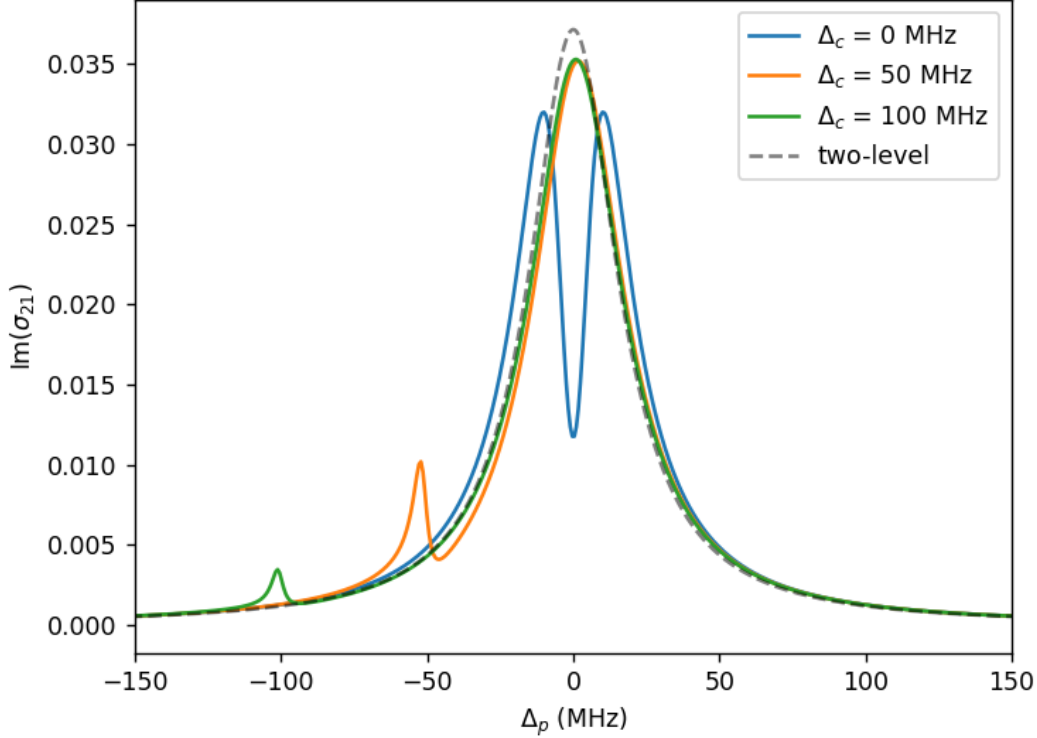


Figure 9: Analytical expression of $\mathcal{I}m(\rho_{21})$ (87) for different detunings of the control laser and in absence of the control beam (dashed black). Rabi frequencies $(\Omega_p, \Omega_c) = (1.36, 18.7)$ MHz, transition linewidth $(\Gamma_{12}, \Gamma_{23}) = (35, 2.2)$ MHz, $\Gamma_{13} = 35$ Hz⁹, and laser linewidth $\gamma_p = \gamma_c = 1$ MHz. $\frac{\Omega_p}{\Omega_c} \approx 0.073$ so that the expression (87) is close to the exact solution, see **E.1 Limit** $\Omega_p \ll \Omega_c, \Gamma_{12}$.

the same expression as in the two-level case (88). So there is effectively no EIT, which is what we expect since the dark state is formed when the two-photon resonance condition (77) is satisfied. We see in Figure 9 that in the case where the control beam is not detuned, the expressions for the simple two-level system and that for EIT are equivalent for $\Delta_p \gg \gamma_{12}$. In the case of large detuning $\Delta_c = 50$ MHz or 100 MHz, the EIT expressions are quite similar to those for the two-level system with a small additional dip at the two-photon resonance (77).

⁹We did not find a tabulated value for the spontaneous decay rate from state 4^1D_2 and 4^1S_1 of neutral calcium, the evaluation to tens of Hz is done in **3.1.2 Neutral calcium atoms**

In the second case $\Delta_p = 0$, we obtain the expression

$$\mathcal{Im}(\rho_{21}) = -\frac{\Omega_p}{2} \frac{1}{\gamma_{12}} \left(1 - \frac{\Omega_c^4}{4} \frac{1}{\gamma_{13}\gamma_{12} + \frac{\Omega_c^2}{4}} \right) \quad (89)$$

We have here a rather different expression than in the two-level case. The imaginary part of ρ_{21} and thus the absorption coefficient is reduced by the factor in parenthesis in the expression (89) which tends to zero under the condition

$$\sqrt{\gamma_{12}\gamma_{13}} \ll \Omega_c. \quad (90)$$

This is what we expect from EIT, as a macroscopic part of the atoms are in the dark state and do not interact with light, the latter propagates without being absorbed by these atoms in the dark state, consequently the absorption coefficient is smaller. In Figure 9, we see that indeed, compared to the two-level case, the EIT curve with a zero detuning of the control beam shows a decrease in absorption.

As in the case of the two-level system, we must also take into account the Doppler shifts effects

$$\Delta_p \rightarrow \Delta_p - \vec{k}_p \cdot \vec{v} \text{ and } \Delta_c + \Delta_p \rightarrow \Delta_c + \Delta_p - (\vec{k}_c + \vec{k}_p) \cdot \vec{v}. \quad (91)$$

Since the general effect of Doppler broadening is to reduce the height of the EIT dip [18], we try to minimize it. In a ladder system, we chose that the two beams propagates in opposite directions in order to limit the effect of the Doppler shifts on the $\Delta_c + \Delta_p$ diagonal element of the hamiltonian (74).¹⁰

When we introduced the dephasing Lindblad operator (28) in the subsection **2.2.3 Specific examples of jump operators**, we detailed different dephasing causes such as the laser linewidth or collisions, as we mentioned that this could significantly affect EIT. In fact, this is true for systems where the linewidth γ_{13} for the two levels forming the dark state, here $|1\rangle$ and $|3\rangle$, is negligible with respect to the linewidth γ_{12} of the transition between the ground state $|1\rangle$ and the intermediate state $|2\rangle$, see the definitions (76). This is particularly true in the case of a Λ system where states $|1\rangle$ and $|3\rangle$ are two sub-levels of the same Zeeman manifold, as the dominant term in γ_{13} is the one corresponding to dephasing effects and will have a strong influence on the condition (90) [18].¹¹

¹⁰For a Λ system, we have to use co-propagating beams in order to minimize the effect of the Doppler shifts [25], because the detuning is $\Delta_c - \Delta_p$.

¹¹In a Λ system the state $|2\rangle$ has a higher energy than the state $|3\rangle$, so that there is no term Γ_{23} in the definition of γ_{13} , on the opposite of the case of a ladder system (76).

Note that we observe EIT for a control beam on resonance $\Delta_c = 0$, approximately when the control Rabi frequency Ω_c is smaller than the typical decay rate of the system like on Figure 9. Since we want to satisfy the condition (90) in the three-level system in neutral calcium atoms and that we have roughly $\Gamma_{23} \approx \frac{\Gamma_{12}}{10}$, see Table 1, we will be above this limit. Hence, we switch to the Autler-Townes splitting regime [26].

2.5.5 Autler-Townes splitting

Autler-Townes splitting (ATS) is similar to electromagnetically induced transparency. The configuration and the experimental setup are the same as in Figure 8. Likewise, the hamiltonian and the Lindblad operator governing the optical Bloch equations are the same as those introduced for EIT respectively (74) and (75). However, ATS occurs when the control field is stronger, roughly when the Rabi frequency of the control field is higher than the transition linewidth between the two states coupled by the probe beam [18]

$$\gamma_{12} \leq \Omega_c. \quad (92)$$

If we continue to consider a control beam on resonance $\Delta_c = 0$, switching to the dressed state picture, a strong enough control beam applied between states $|2\rangle$ and $|3\rangle$ will result in hybridized eigenstates and corresponding eigenenergies [27]

$$|\pm\rangle = \frac{1}{\sqrt{2}} (|2\rangle \pm |3\rangle), \text{ and } E_{\pm} = \pm \frac{\Omega_c}{2}. \quad (93)$$

The condition (92) corresponds to a sufficiently strong control Rabi frequency Ω_c so that the energy levels $E_{\pm}$ with their linewidths do not overlap, as they are farther apart than the linewidth of the absorption signal between states $|1\rangle$ and $|2\rangle$. In this case, the probe beam couples the state $|1\rangle$ with either the state $|+\rangle$ or the state $|-\rangle$ depending on which eigenenergy $E_{\pm}$ (93) its detuning is equal to. The difference with EIT is that there is no more interference but that, depending on the detuning of the probe beam, we form a two-level system, either $\{|1\rangle, |+\rangle\}$ or $\{|1\rangle, |-\rangle\}$. Consequently, on Figure 10 we can see for quite high control Rabi frequencies that the curves look like two Lorentzian respectively centered on the eigenenergies $E_{\pm}$ of the hybridized states (93).

Considering the specific case of the three-level ladder system of the neutral calcium atom $\{4^1S_0, 4^1P_1, 4^1D_2\}$ with the corresponding spontaneous decay rates in Table 1, the transition from the EIT to the ATS regime occurs when the control Rabi frequency is about 17.1 MHz (92) and the condition

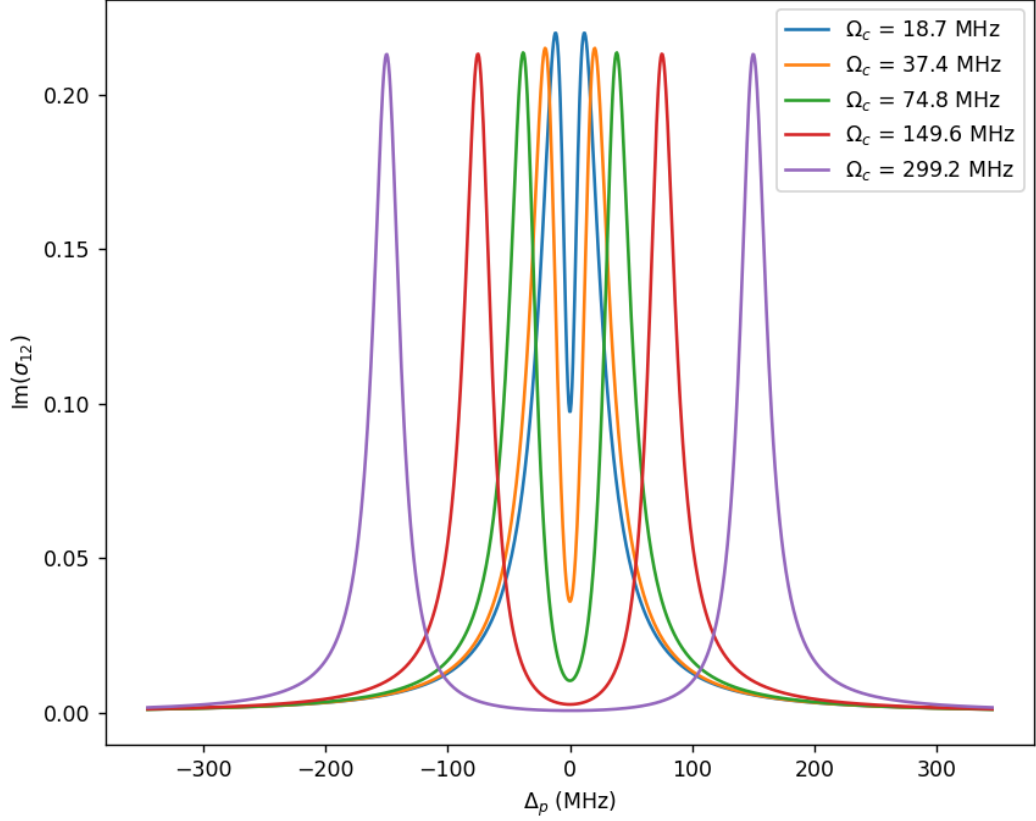


Figure 10: $\mathcal{I}m(\rho_{12})$ for different control Rabi frequencies. Control beam on resonance $\Delta_c = 0$, probe beam Rabi frequency $\Omega_p = 13.6$ MHz, transition linewidth $(\Gamma_{12}, \Gamma_{23}) = (35, 2.2)$ MHz, $\Gamma_{13} = 35$ Hz¹², and laser linewidth $\gamma_p = \gamma_c = 1$ MHz.

to see EIT would be to have the control Rabi frequency much higher than 8.7 MHz (90). The two conditions are drawn under the assumption (86). Therefore, we work in the Autler-Townes splitting regime in the different experiments. Note that it would be interesting to examine precisely how the two conditions defining EIT (90) and ATS (92) change when the ratio of the probe Rabi frequency to the control Rabi frequency is increased.

¹²We did not find a tabulated value for the spontaneous decay rate from state 4^1D_2 and 4^1S_1 of neutral calcium, the evaluation to tens of Hz is done in **3.1.2 Neutral calcium atoms**

2.5.6 Modelling influence of collisions

The data we collected show that the absorption drop does not perfectly match the theoretical expression derived in **E.2 Approximated analytical expression**. Indeed, as we decrease the power of the control beam, the amplitude of the transparency peak tends to decrease faster than expected from the theoretical expression, and if we try to fit the dephasing rates (75) we obtain suspiciously high results of several tens of MHz. One possible explanation is that, as we perform our experiment in an optogalvanic lamp, the effects of the collisions between the calcium atoms and the buffer gas could be more complex than just a simple dephasing process described by the Lindblad operator (75). One possibility would be to try to introduce a collision kernel as did in the article [28], but this would involve more complex optical Bloch equations and thus the fitting of the data would take more time. Therefore, we develop below an empirical model that could explain the observations.

To model this effect mathematically, we draw an analogy with the model of an error in a quantum circuit. A simple example of an error is a bit flip X that occurs with a probability p on a qubit, see [29], and is modeled as

$$\mathcal{U}_{err}(\rho) = (1 - p)\rho + pX\rho X = (1 - p)\mathcal{I}(\rho) + p\mathcal{E}_{bit_flip}(\rho). \quad (94)$$

$\mathcal{I}$ and $\mathcal{E}_{bit_flip}$ are completely positive and trace preserving operators [15], describing a physical evolution of a quantum state. With probability $1 - p$ the state does not undergo a bit flip and evolves under the identity operator $\mathcal{I}$ and in the other case the state evolves under the bit flip operator $\mathcal{E}_{bit_flip}$ defined by its Kraus operator X .

To return to our three-level system, we introduce the completely positive and trace preserving operators describing the evolution of the atoms.

- $\mathcal{I}$ is the identity: $\mathcal{I}(\rho) = \rho$.
- $\mathcal{E}_{2-lvl}$ is the operator which associates to the initial state the steady state solution of the optical Bloch equation for the interaction with the 423 nm laser only: $\mathcal{E}_{2-lvl}(\rho) = \rho^{2-lvl}$.
- $\mathcal{E}_{abs}$ is the operator which associates to the initial state the steady state solution of the optical Bloch equation for the interaction with 423 nm and 733 nm lasers: $\mathcal{E}_{abs}(\rho) = \rho^{abs}$.

The total evolution of our atomic vapor is modeled by the operator

$$\mathcal{E}_{tot} = q\mathcal{I} + (1 - q)[p\mathcal{E}_{2-lvl} + (1 - p)\mathcal{E}_{abs}], \quad (95)$$

where q and p are equivalent to the error probabilities in a quantum circuit (94). q corresponds to the probability that a collision occurring during the excitation from the state $|1\rangle$ to the state $|2\rangle$ prevents this transition from occurring. p is defined in the same way but for the transition between the states $|2\rangle$ and $|3\rangle$. Then, the coherent term ρ_{12} , which defines the absorption of the probe beam in the optogalvanic lamp is

$$\rho_{12} = \mathcal{E}_{tot}(\rho_{ini})_{12} = (1 - q) [p \cdot \rho_{12}^{2-lvl} + (1 - p) \cdot \rho_{12}^{abs}]. \quad (96)$$

The factor $(1 - q)$ can be absorbed in the optical density and defined an effective number of atoms interacting with the beams. Thus, in simulations, we will only introduce a single empirical error probability parameter p to fit the curves.

2.6 Two-color polarization spectroscopy

In this section, we theoretically discuss the two-color polarization spectroscopy technique we use to lock of the 733 nm laser onto the transition between the state 4^1P_1 and the state 4^1D_2 of neutral calcium atoms. It uses the same configuration, atomic levels and lasers, as the experiment of the three-level ladder system and is quite similar to the polarization spectroscopy technique. This is why we named it as such.

2.6.1 Simple analogy with polarization spectroscopy

Before going into the details of the polarization of the light beams and the resulting interaction between the different Zeeman sub-levels of the $\{4^1S_0, 4^1P_1, 4^1D_2\}$ manifolds, we propose a simple introduction to two-color polarization spectroscopy based on the expression of the coherent term ρ_{12} (87). The goal is to gain an overview by considering this simple expression as we did in the case of polarization spectroscopy by first considering a two-level system before taking Zeeman sub-levels into account. We derive this simple expression in the case of the three-level ladder system under the assumption (86).

We assume that the probe beam is locked on resonance $\Delta_p = 0$, so that the expression (87) is now

$$\rho_{12} = i \frac{\Omega_p}{2} \frac{1}{\gamma_{12}} \left(1 - \frac{\Omega_c^2}{4\gamma_{12}} \frac{\gamma_{13} + \frac{\Omega_c^2}{4\gamma_{12}} + i\Delta_c}{\left(\gamma_{13} + \frac{\Omega_c^2}{4\gamma_{12}}\right)^2 + \Delta_c^2} \right). \quad (97)$$

Note that if the control beam is turned off, then the entire parenthesis of the expression (97) is simply equal to 1. Therefore, by chopping the control

beam and using a lock-in amplifier, we can get rid of this constant signal. Then, we have the signal

$$\rho_{12}^{LIA} = i \frac{\Omega_p}{2} \frac{\Omega_c^2}{4\gamma_{12}^2} \frac{\gamma_{13} + \frac{\Omega_c^2}{4\gamma_{12}} + i\Delta_c}{\left(\gamma_{13} + \frac{\Omega_c^2}{4\gamma_{12}}\right)^2 + \Delta_c^2}. \quad (98)$$

This expression (98) has the same dependence on the detuning as the coherent term for the two-level system (30), though with different amplitude and width. Since we exploit this specific detuning dependence in the case of the two-level system to perform polarization spectroscopy and stabilize the 423 nm laser frequency, in an analogous way we can use the detuning dependence of the expression (98) and a setup similar to polarization spectroscopy to lock the 733 nm laser on resonance.

2.6.2 Output signal

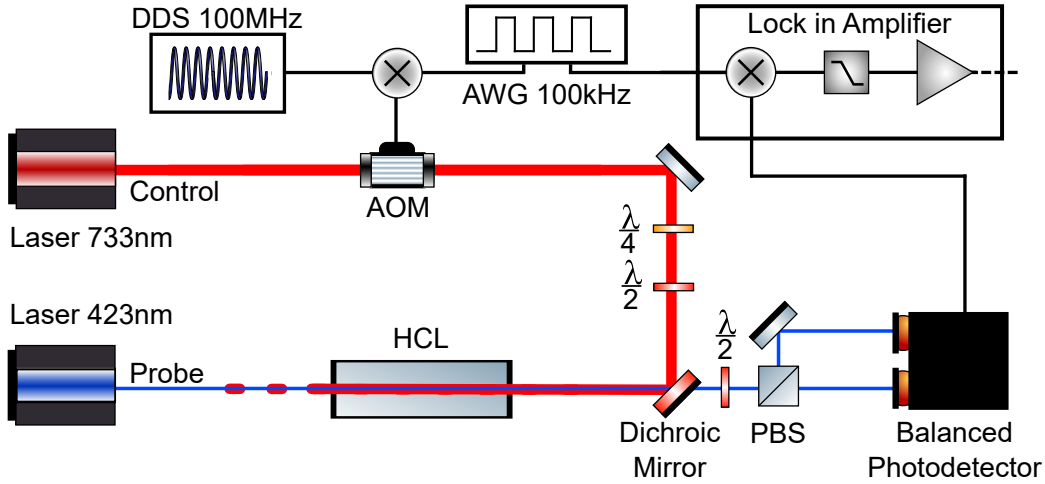


Figure 11: Setup for the two-color polarization spectroscopy¹³. *DDS*: Direct Digital Synthesizer, *AWG*: Arbitrary Waveform Generator, *PBS*: Polarization Beam Splitter, *AOM*: Acousto-Optics Modulator, *HCL*: Hollow Cathode Lamp, $\frac{\lambda}{2}$: half-wave plate, $\frac{\lambda}{4}$: quarter-wave plate.

The two-color polarization spectroscopy setup, see Figure 11, is exactly the same as that of the polarization spectroscopy, see Figure 6, except that the pump beam has been replaced by the 733 nm control beam as in the three-level ladder experiment, see Figure 8. Like in polarization spectroscopy, we

¹³The figure is made with *ComponentLibrary*. *ComponentLibrary* by Alexander Franzen is licensed under a Creative Commons Attribution-NonCommercial 3.0 Unported License.

consider the probe beam to be initially linearly polarized and the control beam to be circularly polarized using the quarter wave plate shown in Figure 11. The presence of the half wave plate just after the quarter wave plate in the path of the control beam serves only to correct any aberrations of the dichroic mirror.

The fact that we are using the same optical elements after the optogalvanic lamp: a half wave plate, a polarization beam splitter, and a balanced photo-detector, as in the polarization spectroscopy setup, implies that the output signal of the balanced photo-detector would also be proportional to the real part of the difference of the refractive indices for the two circularly polarized components of the initially linearly polarized probe beam (65)

$$\Delta I = \frac{E_0^2}{2} e^{-2\text{Im}(n_{tot} + w_{tot}) \frac{\omega}{c} L} \text{Re}(\Delta n) \frac{\omega}{c} L. \quad (99)$$

2.6.3 Polarized light beams and Zeeman sub-levels

Since polarized beams are used, we need to examine the effective Rabi frequencies, (57) and (58), as well as the spontaneous decay rates (60) between the different Zeeman sub-levels.

The Figure 12 summarizes the coefficients for the Rabi frequencies, for linearly¹⁴ (a) and circularly¹⁵ (b) polarized 733 nm laser beam, and the spontaneous decay rates of neutral calcium atom for the Zeeman manifolds 4^1S_0 , 4^1P_1 and 4^1D_2 . The corresponding tables of the Clebsch-Gordan coefficients are given in **Appendix B Clebsch-Gordan Coefficients**.

The general form of the hamiltonian is

$$H = \hbar \left(\sum_i \Delta_i |i\rangle\langle i| + \sum_{i,j} \frac{\Omega_{ji}}{2} |j\rangle\langle i| \right), \quad (100)$$

where i and j are the different Zeeman sub-levels, Δ_i is the corresponding detuning, and Ω_{ji} is the corresponding Rabi frequency, see equations (57) and (58) depending on the polarization of the beams. The Linbald operators for spontaneous decays are of the form

$$\mathcal{L}_{sp} = -\frac{1}{2} \left(\sum_{(i,j)} \Gamma_{ij} \left\{ \sum_k \rho_{kj} |k\rangle\langle j| + \rho_{jk} |j\rangle\langle k| - 2\rho_{jj} |i\rangle\langle i| \right\} \right), \quad (101)$$

where (i, j) corresponds to a possible decay channel from state j to state i , and Γ_{ij} is the corresponding decay rate (60). For simplicity, we do not

¹⁴We consider a linear polarization along the x axis, as in **2.3.1 Rabi frequency**

¹⁵We consider a circular right polarization, as in **2.3.2 Spontaneous decay rate**

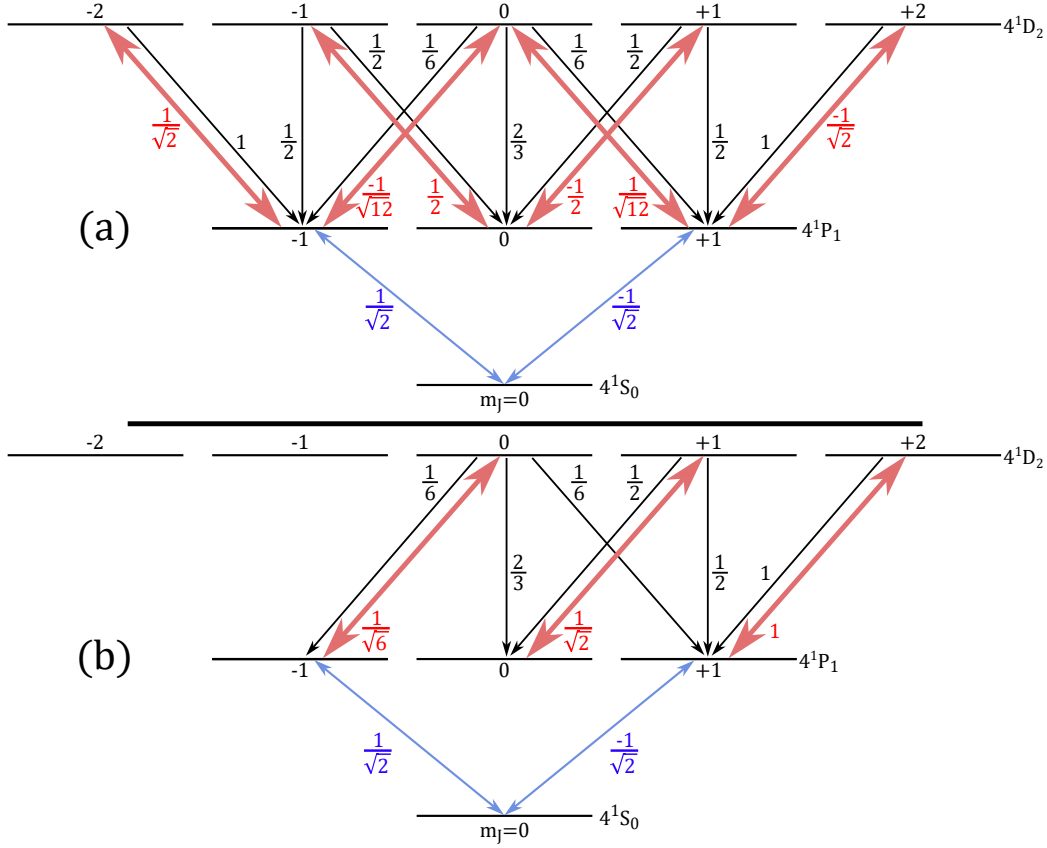


Figure 12: Coupling of Zeeman sub-levels in the two-color polarization spectroscopy setup for 733 nm laser (a) linearly and (b) circularly polarized. The numbers correspond to the coefficients multiplying the Rabi frequencies and the spontaneous decay rate between P and D levels

consider dephasing Linbald operators for laser linewidth or collisions. We do not develop an analytical expression, like in the three-level ladder system, see **E.2 Approximated analytical expression**, because there are too many levels. Instead, we directly perform simulations with the steady state solver of *QuTip* [30].

Eventually, we can quickly analyze the systems presented in Figures 12 (a) and (b) from a symmetry point of view. Indeed, when the control beam is linearly polarized, we see in Figure 12 (a) that the coefficients applied to the Rabi frequencies are antisymmetric and those applied to the spontaneous decay rates are symmetric with respect to the Zeeman sub-levels. These

symmetries imply symmetric expressions for the level population,

$$\rho_{m_j, m_j} = \rho_{-m_j, -m_j}, \quad (102)$$

and anti-symmetric expressions for the coherent terms,

$$\rho_{m_j, m_i} = -\rho_{-m_j, -m_i}. \quad (103)$$

We can be convinced of this statement by examining the dependence of the steady state coefficients of the density matrix for the two-level system (30) on the sign of the Rabi frequency. Due to this symmetric behaviour, (102) and (103), we expect the refractive indices for the two circularly polarized components of the linearly polarized probe beam to be the same, so that the output signal (99) is zero.

Compared to the linear polarization case, the symmetry is broken when the control beam is circularly polarized, see Figure 12 (b). We expect the refractive indices of the two components of the linearly polarized probe beam to be different, such that the output signal (99) is non-zero.

2.6.4 Difference of refractive indices

We now know that the output signal of the balanced photo-detector is proportional to the real part of the difference of the refractive indices (99). To determine its expression, we can follow the same steps as those detailed in the subsections **2.2.6 Evolution of a monochromatic wave in atomic medium** and **2.5.3 Absorption coefficient of a three-level ladder system**. The dielectric polarization, considering only the components oscillating at the probe beam frequency ω_p is

$$\vec{P}_p^{(+)} = \epsilon_0 \chi E_0 \frac{(\vec{\epsilon}_- - \vec{\epsilon}_+)}{\sqrt{2}} e^{-i\omega_p t} \approx \frac{N}{V} d_{12} (\rho_{-1,0} \vec{\epsilon}_- + \rho_{+1,0} \vec{\epsilon}_+) e^{-i\omega_p t}, \quad (104)$$

where $\rho_{\pm 1,0} = \langle J = 1, m_j = \pm 1 | \rho | 0, 0 \rangle$.¹⁶ From the expression (104), we derive the linear dielectric susceptibility for the two circularly polarized components of the linearly polarized probe beam

$$\chi_{\pm} = -\frac{N}{V} \frac{|d_{12}|^2}{\hbar \epsilon_0} \frac{\rho_{\pm 1,0}}{\sqrt{2} \Omega_{\pm 1,0}} \quad \text{with} \quad \Omega_{\pm 1,0} = \mp \frac{\Omega_p}{\sqrt{2}}. \quad (105)$$

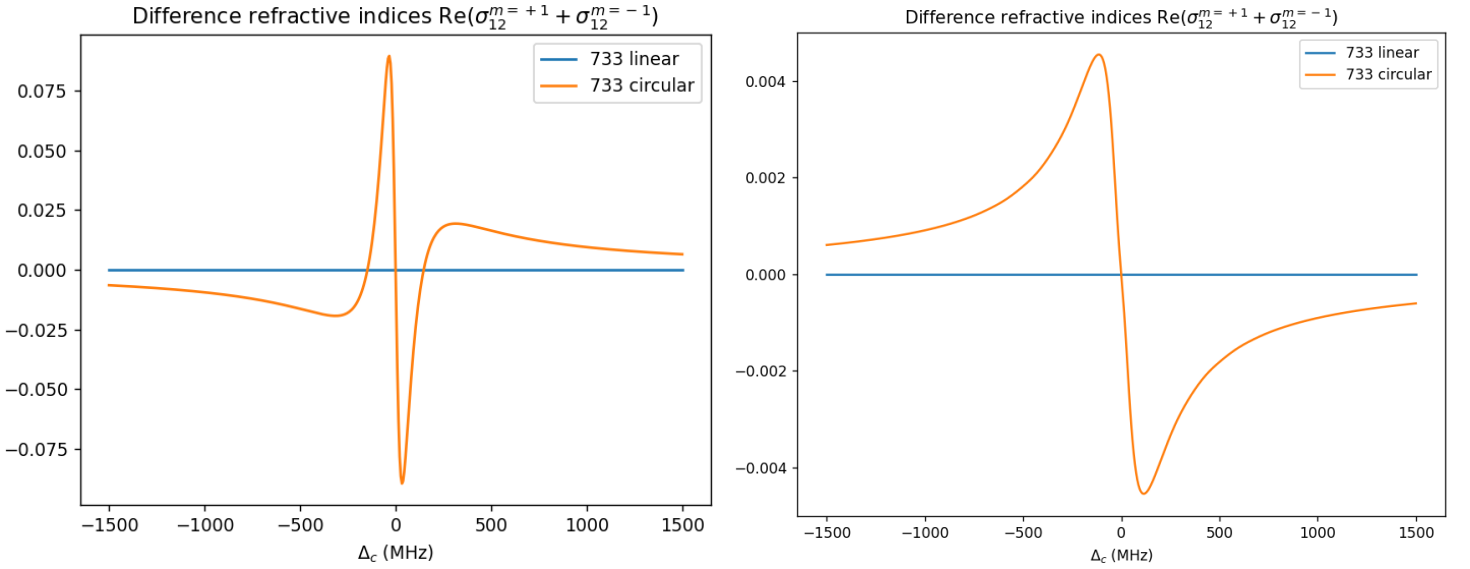
¹⁶Note that we neglect the steady state term $\rho_{m_P=0, m_S=0}$ corresponding to a component along the z axis for the dielectric polarization. As there is no direct coupling with Rabi frequencies from the ground state to this level the resulting steady state coherent term value should be small, if not zero.

We assume that the probe beam is sufficiently weak, so that we can replace the atomic density difference $\frac{\Delta N}{V}$ with the simple atomic density $\frac{\Delta N}{V}$ in the expression for the refractive index (42).

Due to opposite sign of the Rabi frequencies $\Omega_{\pm 1,0}$, the difference of refractive indices is

$$\Delta n = \frac{1}{2} \Delta \chi = \frac{1}{2} \frac{N}{V} \frac{|d_{12}|^2}{\hbar \epsilon_0} \left(\frac{\rho_{-1,0}}{\sqrt{2} \Omega_{-1,0}} - \frac{\rho_{+1,0}}{\sqrt{2} \Omega_{+1,0}} \right) = \frac{1}{2} \frac{N}{V} \frac{|d_{12}|^2}{\hbar \epsilon_0} \frac{\rho_{+1,0} + \rho_{-1,0}}{\Omega_p} \quad (106)$$

Then the signal (99) is proportional to the sum of the steady state solu-



(a) Difference of the coherent terms without Doppler shift (b) Difference of the coherent terms with Doppler shift

Figure 13: Real part of the difference of the coherent terms, which is proportional to the real part of the difference of refractive indices, without (a) and with (b) Doppler shift at a temperature of 400 K. Rabi frequencies $(\Omega_p, \Omega_c) = (40, 150)$ MHz, the dephasing rates are $\gamma_p = \gamma_c = 1$ MHz, the spontaneous decay rates are $(\Gamma_{12}, \Gamma_{23}) = (35, 2.2)$ MHz, $\Gamma_{13} = 35$ Hz¹⁷, and the detuning of the probe laser is $\Delta_p = 0$. Simulations are run with *QuTip* [30]

tions of the coherent terms between the ground state $|0, 0\rangle$ and the states $|1, -1\rangle$ and $|1, +1\rangle$. Combined with the antisymmetry argument (103) and the general expression for the output signal (99), it is easy to see that the

¹⁷We did not find a tabulated value for the spontaneous decay rate from state 4^1D_2 and 4^1S_1 of neutral calcium, the evaluation to tens of Hz is done in **3.1.2 Neutral calcium atoms**

resulting signal in the case of a linearly polarized control beam is zero, which is effectively the result of simulations in Figure 13. In addition, we have to take into account the Doppler broadening. Indeed, we are in a ladder configuration and the wavelengths of the lasers are $\lambda_p = 423$ nm and $\lambda_c = 733$ nm. The effective wavevector $\vec{k}_{eff} = \vec{k}_p - \vec{k}_c$ is therefore non zero and the Doppler shifts $\vec{k}_{eff} \cdot \vec{v}$ are significant, i.e. $k_{eff}v_{th} \gg \gamma_{ij}$. To take into account the different Zeeman sub-levels as well as the Doppler broadening, we use the steady state solver of *QuTip* [30]. Simulations of the real part of the sum of the two coherent terms in the refractive index expression (106) yield a dispersion-like signal as shown in Figure 13 (a), which is broaden and damped as we take into account the Doppler shifts in Figure 13 (b). The signal in Figure 13 (a) is mainly due to the different control Rabi frequencies coupling the states $|J = 1, m_J = -1\rangle$ and $|J = 1, m_J = +1\rangle$ to the manifold 4^1D_2 , with $\frac{1}{\sqrt{6}}$ and 1 factors, respectively, as indicated in Figure 12 (b). In Figure 13 (b), the center of the dispersion-like signal verifies the two-photon resonance condition (77). So if the 423 nm is already locked to transition between the state 4^1S_0 and the state 4^1P_1 , we can use the signal in Figure 13 (b) to lock the 733 nm laser on the $4^1P_1 \leftrightarrow 4^1D_2$ transition.

3 Experimental Apparatus

3.1 Atomic source

The atomic source we use in the different experiments is an optogalvanic lamp, also called a see through hollow cathode lamp. It allows to obtain an atomic vapor of neutral atoms, in our case calcium, and to shine lasers through it, which is well adapted for spectroscopy.

3.1.1 Optogalvanic lamp

In our various experiments, the atomic sources we use are optogalvanic lamps, from *SpectroLamps*, for calcium atoms. The working principle of the hollow cathode lamp light sources is detailed in [31] which is our main reference in this subsection. The optogalvanic lamp has the same operating principle as a hollow cathode lamp, although it is designed to be used as a reference for locking lasers, see [32].

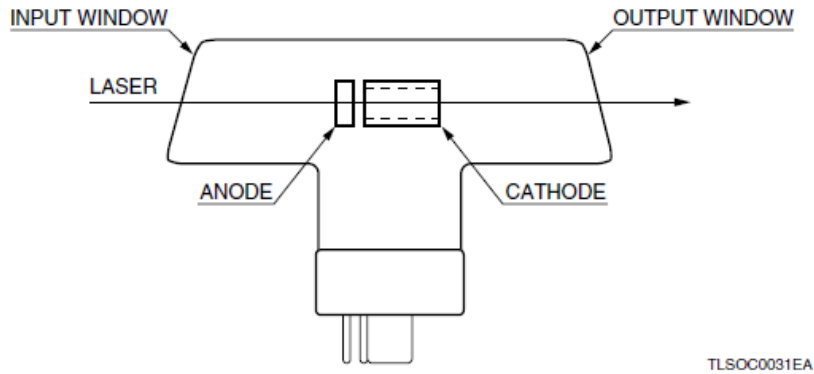


Figure 14: Drawing of a optogalvanic lamp taken from [32]

As shown in Figure 14, the see through hollow cathode lamp consists of two electrodes. The cathode is coated with the atomic species of interest, in our case calcium. The glass envelope contains a buffer gas which is a noble element, usually neon or argon. This buffer gas is ionized when a potential is applied between the two electrodes. The ion nuclei will then be attracted by the cathode and when they reach the cathode, they collide with the calcium layer and neutral atoms are sputtered.

The buffer gas used is always a noble gas so that it does not form molecules that would otherwise have rotating and vibrating modes. These modes would

generate a broad spectrum that would probably interfere with the one of the species of interest.

To control the lamp, we can change the current intensity applied to it. We need to find a balanced value for the current. Indeed, on the one hand, the higher the current, the more atoms of the species of interest are emitted and therefore the higher the signal intensity. On the other hand, the higher the current, the higher the temperature of the atoms, which implies more significant Doppler broadening effect. In addition, a higher current also equates to a shorter lifetime for the hollow cathode lamp, as the emission rate of the atoms is higher. Besides, the lamp is manufactured to operate for a certain amount of time at up to 5 mA current according to its manufacturer *SpectroLamps*.

3.1.2 Neutral calcium atoms

With the optogalvanic lamp, we obtain a vapor of neutral calcium atoms mainly in the ground state [33]. Calcium atoms are alkaline earth metals of atomic number $Z = 20$, so their ground state electronic structure is $[Ar](4s)^2$ or 4^1S_0 . The excited electronic states that are of interest in the experiments are $[Ar](4s)^1(4p)^1$ or 4^1P_1 separated from the ground state by 423 nm and $[Ar](4s)^1(4d)^1$ or 4^1D_2 separated from 4^1P_1 by 733 nm, see Figure 15.

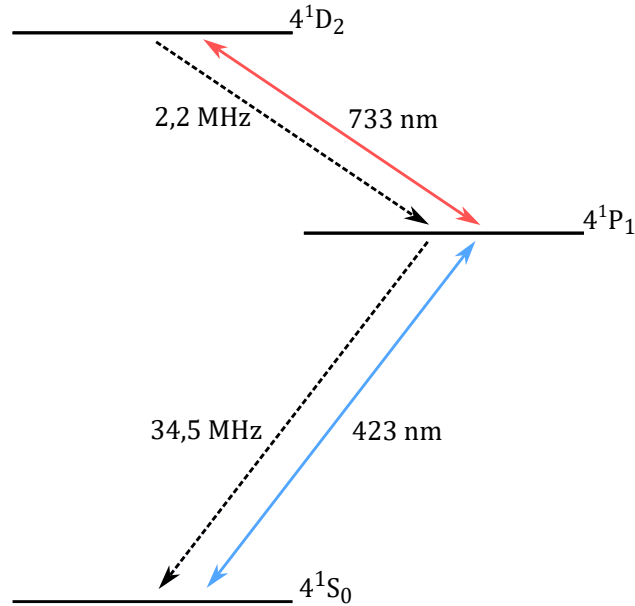


Figure 15: Structure of neutral calcium atom

In Figure 15, the transition wavelengths are indicated in plain arrow and

the decay rates by the dotted lines, their values are given in Table 1. No references was found for the decay rate for the quadrupole transition from the state 4^1D_2 to the state 4^1S_0 . However, we can get a rough estimate because the decay rate of a quadrupole transition is approximately [34]

$$\Gamma_{e \rightarrow g}^{QP} \approx \frac{\omega_0^5 |\langle g|Q_e|e \rangle|^2}{\pi \hbar \epsilon_0 c^5} \quad \text{with} \quad |\langle g|Q_e|e \rangle| = \frac{1}{2} e \langle \vec{r}^2 \rangle \approx \frac{e}{2} \left(a_0 \frac{n^2}{Z} \right)^2. \quad (107)$$

n is the principal quantum number of the outermost electron of the state 4^1D_2 , Z is the number of protons and a_0 is the Bohr radius. Using the expression (107), the quadrupole decay rate from the state 4^1D_2 to the state 4^1S_0 is of the order of tens of hertz. Furthermore, there is a possibility for atoms in the 4^1P_1 state to decay into a different state 3^1D_2 , but the associated decay rate is 0.35 kHz [17] so we neglect it.

3.2 Ti:Sapphire *MSQUARED SolsTiS* laser

The 733 nm laser source we want to lock onto the $4^1P_1 \leftrightarrow 4^1D_2$ transition of neutral calcium atoms is a Ti:Sapphire *MSQUARED SolsTiS* laser with a narrow tunable linewidth. In order to be able to compare the stability of this laser with internal locking techniques and two-color polarization spectroscopy, we give here a quick description of the laser based on its user manual [12].

On Figure16, we can see the main elements constituting the Ti:Sapphire laser.

- The *Ti:S Rod* is the laser medium. It defines the gain envelope, i.e., for a specific frequency component, the factor by which the beam is amplified as it passes through the Ti:Sapphire medium.
- The *optical diode* imposes the direction of light propagation within the bow-tie cavity. It works in a similar way to a Faraday isolator [35].
- The piezo-mounted mirror M_3 can modify the length of the bow-tie cavity and hence change the frequency of the longitudinal modes.
- The *birefringent filter* selects the wavelength of the light oscillating in the cavity. This selection is narrower than the width of the gain envelope of the Ti:Sapphire medium, although it is still quite large.¹⁸
- The *etalon* acts similarly to a Fabry-Pérot cavity and provides a more accurate selection of the wavelength of the light beam [36].

¹⁸There is no detailed number about the width of the different optical components.

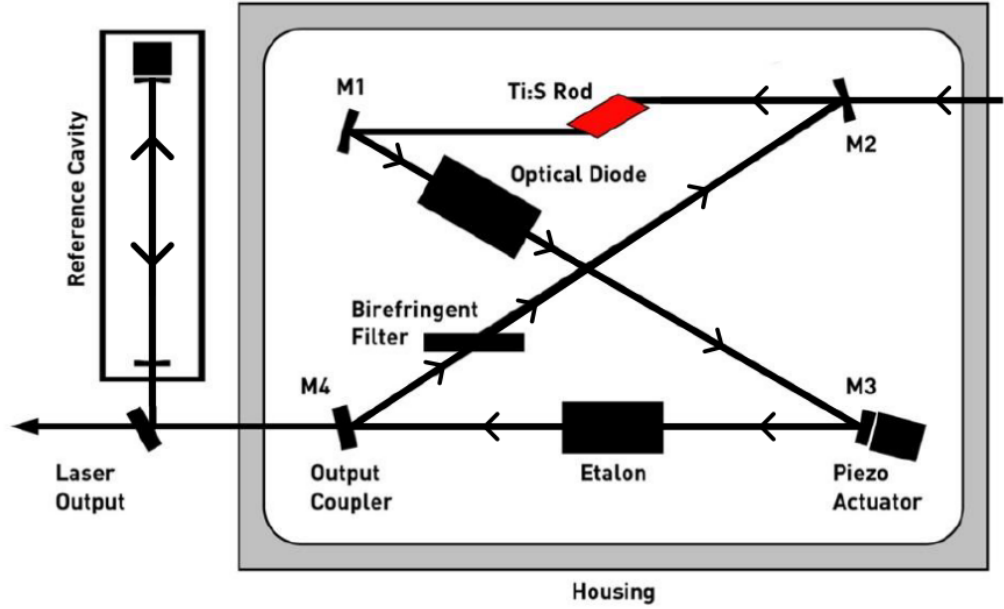


Figure 16: Plan of the Ti:Sapphire laser, adapted from [12]. The mirrors of the bow-tie cavity are labeled M_i .

- In the *SolsTiS - SRX* Ti:Sapphire laser model we use, there is also an additional reference cavity after the bow-tie cavity that can be used to feedback to the piezo-mounted mirror M_3 to have a fine tuning and good frequency stability of the laser.

There are two locking options for this Ti:Sapphire *M SQUARED SolsTiS* laser. The first is to use the *étalon* whose length can be controlled to stabilize the frequency. Second, the reference cavity can be used to control the piezo-mounted mirror M_3 to obtain an even more accurate control of the laser frequency.¹⁹

3.3 *HighFinesse WS6 - 200 Series* wavemeter

The goal of this master's thesis is to stabilize the frequency of the 423 nm and 733 nm lasers. To be able to quantify their stability we use a *HighFinesse WS6 - 200 Series* wavemeter. For technical reasons, we use two different

¹⁹Using the reference cavity, the user manual announces a linewidth of about 50 kHz calculated over 100 μ s. Besides, the user manual of the Ti:Sapphire announces that the laser in free operation has a frequency stability of about 100 MHz/hours/K [12].

wavemeters, although they are the same models, in two separate laboratories called B20 and B25. The wavemeter recording the frequency of the 423nm laser is the one in B25, and the wavemeter recording the frequency of the 733nm laser is the one in B20.

On the data sheet of the *HighFinesse WS6 - 200 Series* wavemeter [37], there are two main characteristics that influence the frequency measurement. The first one is the absolute precision of 200 MHz of the wavemeter. It corresponds to the proximity of the average frequency it measures to the actual value. If we tune the lasers according to the wavemeter then we could be up to 200 MHz detuned from what we expect, though by using a spectroscopy technique we get rid of this problem. The second characteristic is the standard deviation of 4 MHz. It means that even for a locked laser whose actual frequency standard deviation is less than this limit, the standard deviation of the frequency measurement that the wavemeter would return would be about 4 MHz.

This announced standard deviation of 4 MHz is in principle too large in our case, because to achieve the two-color cooling scheme we would like to stabilize the lasers frequency below 1 MHz. To check what is the actual standard deviation of this wavemeter model, we monitor the frequency with the wavemeter in B20 of a 729 nm laser locked with a high finesse cavity with a frequency standard deviation below 5 kHz [38], so that most of the signal noise is caused by the wavemeter. It turns out that the standard deviation calculated over 5 minutes is about 0.4 – 0.6 MHz, see subsection **4.3.2 Locking of the 733 nm laser**. Consequently, as this standard deviation is below the 1 MHz limit we want to reach we can use these wavemeters to monitor the frequency of both lasers.²⁰

3.4 423 nm Polarization spectroscopy

The experimental setup for the polarization spectroscopy is shown in Figure 17 which corresponds to the description in Figure 6. The optical components are labeled in white from 1 to 9.

- The 423 nm laser beam comes directly from a *Toptica DL pro* laser.
- 1 is a half-wave plate, which coupled to the polarization beam splitter 2, allows adjustment of the amount of power for the probe and the pump beams.

²⁰Note that the standard deviation of the wavemeter in B25 laboratory could be different from the one in B20 laboratory.

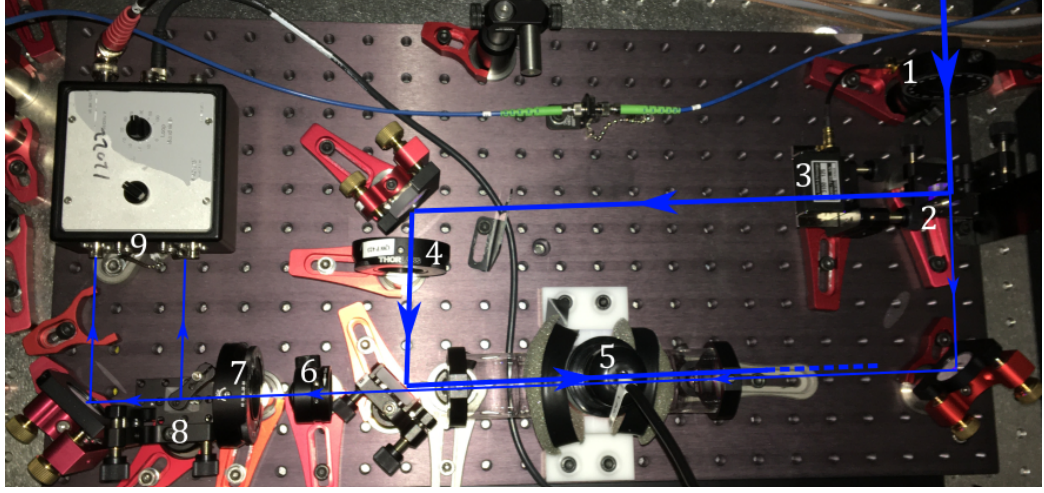


Figure 17: Experimental setup of the polarization spectroscopy in an optogalvanic lamp with neutral calcium atoms

- 3 is the acousto-optic modulator, a *Gooch & Housego* model 3100-125. We use it as a chopper, the 0 order being blocked and the -1 order being used as the pump beam. It is supplied by a 100 MHz amplified RF signal from a DDS board and modulated via a switch, a *Mini-circuits ZASWA-2-50DR+*, and a square wave signal from an arbitrary waveform generator, a *Keysight 33500B Series*, at 100 kHz. The waveform generator also feeds the lock-in amplifier to achieve the same internal frequency as the signal.
- 4 is a quarter wave plate that transforms the linearly polarized pump beam into a circularly polarized beam.
- 5 is an optogalvanic lamp, from *SpectroLamps*, containing the neutral atoms of calcium. The current is regulated with an analogical current controller. In the lamp, the pump and the probe beams must be as much superposed as possible.
- 6 is an iris to avoid reflections of the pump beam on the windows of the optogalvanic lamp to illuminate the photo-detector.
- 7 is the half wave plate to correct the difference of refractive indices induced by the windows of the see through hollow cathode lamp and to rotate the polarization of the probe beam so that the final signal has the correct expression (65).

- 8 is a polarization beam splitter, which projects the electromagnetic field onto each of the x and y axes. Therefore, the balanced photo-detector 9, a *Nirvana detector* model 2007, returns the difference of intensity $I_x - I_y$.

Then, the output signal of the balanced photo-detector goes to the lock-in amplifier, a model *SR830* from *Stanford Research Systems*, to be demodulated, to filter the noise at frequencies different from 100 kHz and to be eventually amplified.

3.5 Absorption drop in a three-level system

The experimental setup for observing the absorption drop in the three-level ladder system $\{4^1S_0, 4^1P_1, 4^1D_2\}$ of neutral calcium atoms is shown in Figure 18, which corresponds to the description in Figure 8 (b). The optical parts are labeled in white from 1 to 5 and correspond to the description made below.

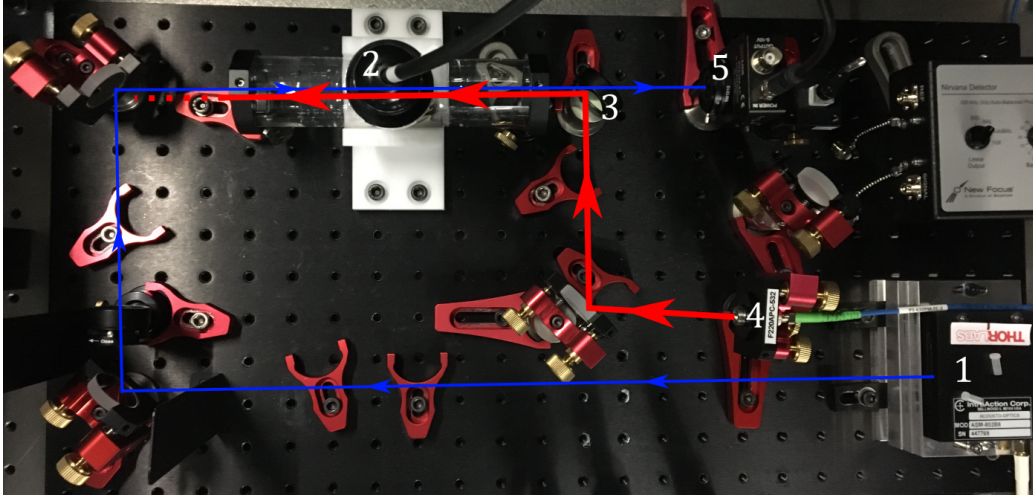


Figure 18: Experimental setup for the three-level ladder system in a calcium optogalvanic lamp.

- The 423 nm laser beam is coming from the *Toptica DL pro* laser and the 733 nm laser is from a narrow linewidth, tunable Ti:Sapphire *M SQUARED SolsTiS* laser.
- 1 is an 80 MHz acousto-optic modulator, from *IntraAction Corp.* model *ASM-802B8*, modulated via a switch, a *Mini-circuits ZASWA-2-50DR+*,

and a waveform generator, *Keysight 33500B Series*, at 100 kHz. This acousto-optic modulator and the block wall allow only the +1 order of the 423 nm beam to pass to the optogalvanic lamp and to the photo-detector.

- 2 is the optogalvanic lamp, from *SpectroLamps*, containing the neutral atoms of calcium. The current is regulated with a high voltage power supply *Model PS310* from *Stanford Research Systems*.
- 3 is the dichroic mirror transmitting the 423 nm blue light and reflecting the 733 nm red control beam coming from the optical fiber 4.
- Eventually, the 423 nm probe beam goes onto the photo-detector 5, a *Thorlabs* model *PDA36A-EC*.

The resulting output signal is then filtered through a lock-in amplifier, model *SR830* from *Stanford Research Systems*. The reference signal for the lock-in amplifier is created by the same waveform generator signal used to modulate the acousto-optic modulator.

3.6 423 nm - 733 nm Two-color polarization spectroscopy

The experimental setup to lock the 733 nm laser, based on two-color polarization spectroscopy, is shown in Figure 19, which corresponds to the description in Figure 11. The optical devices are labeled in white from 1 to 8.

- As in the three-level ladder system setup, the 423 nm laser beam is coming from the *Toptica DL pro* laser and the 733 nm laser is from a narrow linewidth, tunable Ti:Sapphire *M SQUARED SolsTiS* laser.
- 1 is an 80 MHz acousto-optic modulator, from *IntraAction Corp.* model *AOM-802B8*, which is powered by a DC RF signal, so that the 423 nm beam going to the experiment is the +1 order coming out of the acousto-optic modulator. This acousto-optic modulator is of no use in this setup but is needed if we want to return to the Autler-Townes splitting setup described in the previous subsection, to chop the 423 nm probe beam.
- 2 is the optogalvanic lamp, from *SpectroLamps*, containing the neutral calcium atoms. The current is set with a high voltage power supply *Model PS310* from *Stanford Research Systems*.

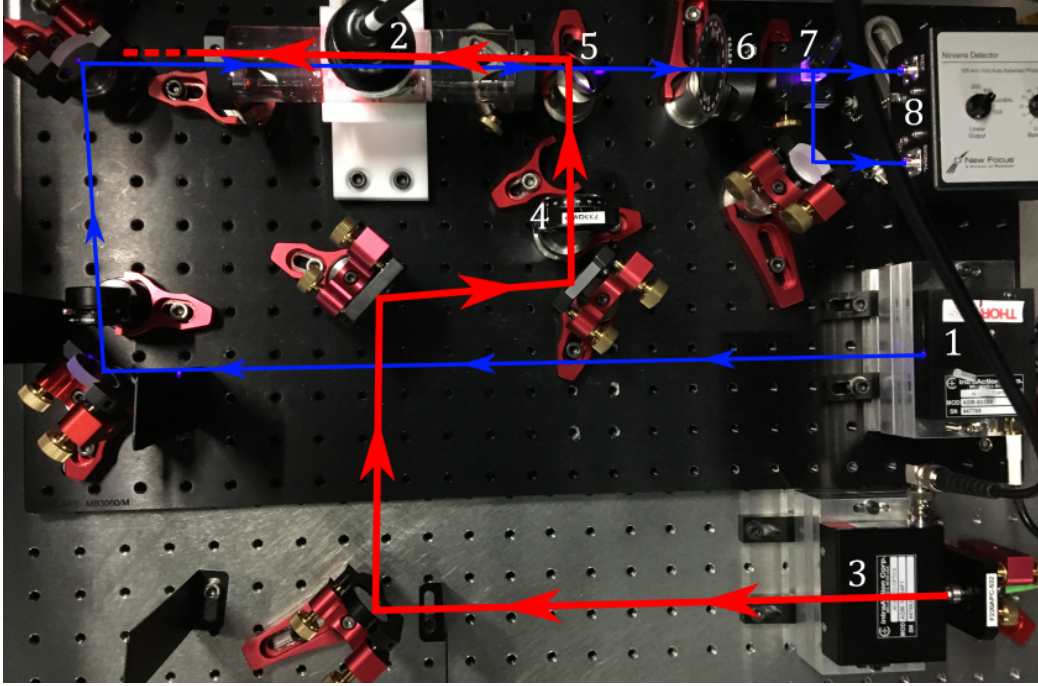


Figure 19: Experimental setup for the two-color polarization spectroscopy technique in a calcium optogalvanic lamp.

- 3 is a 100 MHz acousto-optic modulator, from *IntraAction Corp.* model *AOM-1002AF1*²¹, which is modulated by a switch, a *Mini-circuits ZASWA-2-50DR+*, and a 100 kHz square wave signal from a *Keysight 33500B Series* waveform generator. The latter also feeds the lock-in amplifier, model *SR830* from *Stanford Research Systems*, which is used to filter noise from the signal. With the block wall, only the -1 order of the beam is sent to the experiment.
- 4 is the quarter wave plate mentioned in Figure 11 that is used to change the initial linear polarization of the 733 nm beam to a circular polarization.
- 5 is the dichroic mirror transmitting the 423 nm beam and reflecting the 733 nm control beam.
- 6 is the half wave plate allowing to correct the difference of refractive indices induced by the windows of the optogalvanic lamp. This half

²¹The acousto-optic modulator *AOM-1002AF1* is designed for a laser light at 630 nm wavelength, though it was the closest one we found [39]. The resulting diffraction efficiency for the 733 nm laser beam is about 40 % before chopping.

wave plate also rotates the polarization of the 423 nm probe beam, so that once it passes through the polarization beam splitter 7 and is detected in the balanced photo-detector 8, model *2007* from *Nirvana detector*, the signal is proportional to the real part of the difference of refractive indices (99).

4 Results

4.1 423 nm Polarization spectroscopy

In this section, we present our results for polarization spectroscopy with the 423 nm laser light and the transition of neutral calcium atoms between the states 4^1S_0 and 4^1P_1 . We first examine the dispersion-like signal we obtain with the setup, and then we attempt to characterize the frequency stability of the laser when we use the polarization spectroscopy technique to lock it.

4.1.1 Polarization spectroscopy signal

The dispersion-like signals shown in Figure 20 (a) and (b) are recorded on an oscilloscope, model *TBS 1104* from *Tektronix*. To transform the time scale into a frequency scale, we calibrate the voltage provided by the *EVIL* PI-controller [40] to the laser frequency using the wavemeter²². In the end, using the slope in voltage per unit time of the ramp signal applied with the *EVIL* PI-controller, we get a coefficient in frequency per unit time and we convert the oscilloscope time axis to a frequency axis by multiplying the recorded time by this coefficient, albeit with an arbitrary frequency offset.

We collect the data with the experimental setup shown in Figure 17 for the fixed parameters:

- The probe beam power is 0.12 mW.
- The radii of the probe and the pump beam are 0.46 mm, according to the gaussian fit realized with the beam profiler from *Thorlabs*.
- The time constant of the lock-in amplifier is 300 μ s.
- The sensitivity of the lock-in amplifier is 2 mV.

Then we change the intensity applied to the optogalvanic lamp and for each intensity we take seven different values for the pump beam power. These values are tabulated in Table 2.

First, it should be noted that the current intensities in the polarization spectroscopy setup are set with an analog current controller, so the error on the current intensity can be estimated to be about 0.5 mA. Second, the beams powers are measured in front of the optogalvanic lamp windows. To

²²The wavemeter in the B25 laboratory see **3.3 *HighFinesse WS6 - 200 Series* wavemeter**.

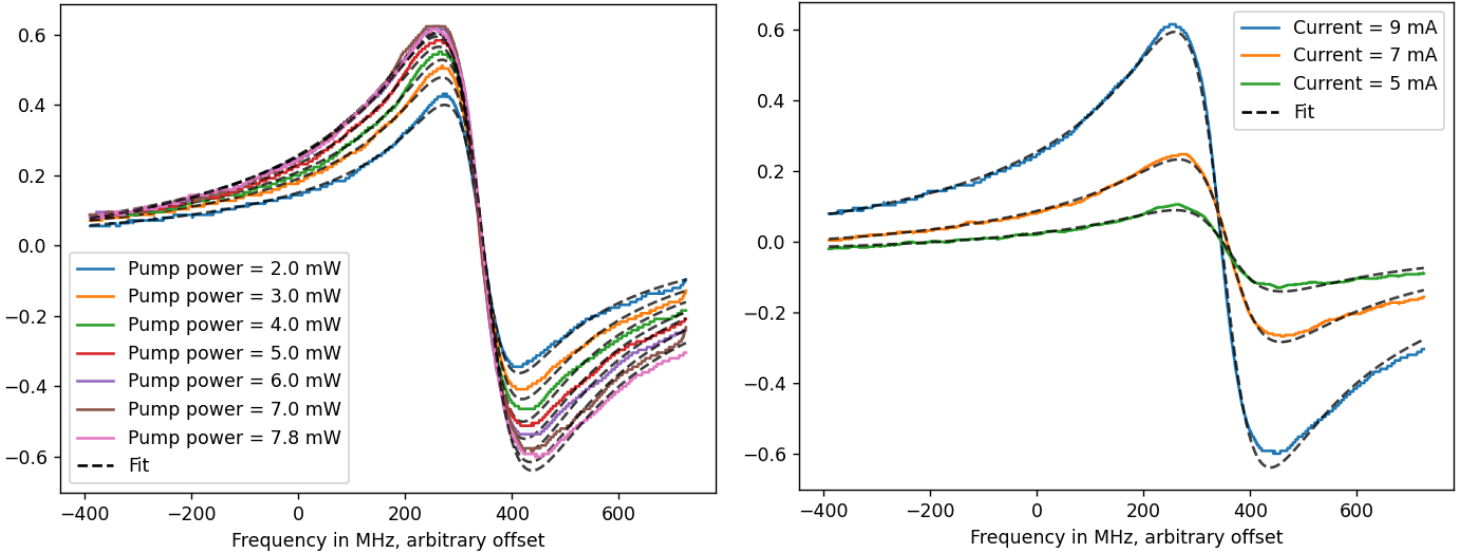
Intensity in mA	5			7		9	
Pump beam power in mW	2.0	3.0	4.0	5.0	6.0	7.0	7.8

Table 2: Parameters to get polarization spectroscopy signal. The current intensities are those applied to the optogalvanic lamp. The power is measured in front of the lamp.²³

be more precise, we calculate the transmission coefficient of the intensity T_1 for one window of the lamp cell. To do this, we turn off the lamp and measure the square root of the ratio between the output and the input power as the beam passes through the lamp. This transmission coefficient is not negligible and is evaluated over five measurements to

$$T_1 = 0.809 \pm 0.003. \quad (108)$$

We use this value in our fits to determine the Rabi frequencies of the two beams in the atomic medium.



(a) Polarization spectroscopy signals for different pump beam power and a current intensity of 9 mA (b) Polarization spectroscopy signals for different current intensities and a 7.8 mW pump power

Figure 20: Polarization spectroscopy curves (colored lines) and their fit (dashed black lines) with the expression for the polarization spectroscopy signal (67).

²³In fact, with the powermeter we measure half of these values for the pump beam power as it is a chopped signal.

For each current, we fit the curves with the theoretical expression (67) derived in subsection **2.4.2 Polarization spectroscopy**, where we take into account the transmission coefficient of the intensity (108). In Figure 20 (a), the seven curves, corresponding to the seven values of pump beam power and a current intensity of 9 mA applied to the optogalvanic lamp, are fitted with the following parameters: a temperature, a dephasing rate, an amplitude factor, a center frequency, and an offset, although the latter parameter should be close to zero.

In Figure 20 (b), we plot the polarization spectroscopy signal for different intensities applied to the optogalvanic lamp and for the same pump beam power of 7.8 mW. We can see that the higher the current intensity, the higher the signal amplitude and the steeper the slope around zero. This is logical because the signal is proportional to the density of neutral calcium atoms and that a higher intensity implies a higher atomic density. Also, since polarization spectroscopy is Doppler free, a higher temperature would not affect the width of the signal. Thus, increasing the intensity applied to the optogalvanic lamp can be a simple and effective way to improve the frequency stability when locking the laser with the polarization spectroscopy.

The seven curves associated with the same current intensity, like in Figure 20 (a), should have the same temperature and the same dephasing rate due to collisions and the laser linewidth. This is because a fixed current intensity applied to the lamp determines the temperature and the velocity distribution and thus the collision rate in the lamp. First, concerning the fitted temperatures, the mean values and the standard deviations for each current intensity applied to the optogalvanic lamp are reported in Table 3. The standard deviations for the fitted temperatures in Table 3 are signifi-

		Current intensity		
		5 mA	7 mA	9 mA
Temperature	Mean Value	250	279	421
in K	Standard deviation	62	49	73

Table 3: Mean values and standard deviation of the seven fitted temperatures for the seven pump power and a given current intensity, see Table 2.

cant, although these values for the 7 mA and 9 mA current intensities are consistent with those we derive more correctly in a next subsection: **4.2.3 Determining the temperatures**, when we examine the drop of absorption in the three-level ladder system of neutral calcium atoms.

The fitted dephasing rates are plotted in Figure 21, they clearly show a decreasing behaviour as a function of the pump beam power. We do not

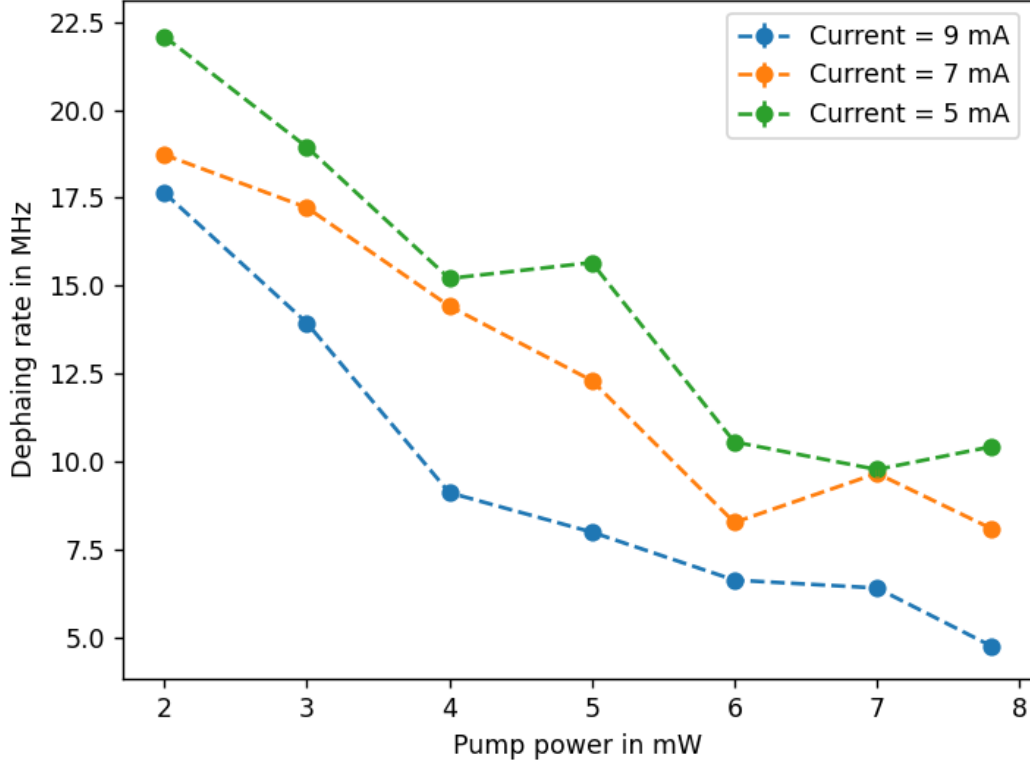


Figure 21: Fitted dephasing rate as a function of the pump beam power for the different current intensities applied to the optogalvanic lamp.

know how to explain this behaviour, it could be related to what we observe in the absorption drop in the three-level system and forced us to introduce an empirical error probability, see **2.5.6 Modelling the influence of collisions**, to be able to fit the data and which turn out to be dependent on the 733 nm control beam power. Note that the dephasing rates seem to converge for high pump powers to values of a few MHz which are the typical values we expect.

4.1.2 Locking of the 423 nm laser

We use the polarization spectroscopy dispersion-like signal, as in Figure 20, to lock the 423 nm laser of the neutral calcium atom transition between the state 4^1S_0 and the state 4^1P_1 . The dispersion-like signal is sent to the *EVIL* PI-controller [40], then the *DevilClient* software allows us to control the PI parameters of the control loop. The corrected voltage is then fed back to the piezoelectric controller of the 423 nm *Toptica DL pro* laser.

The results of the locking of the 423 nm laser are shown in Figure 22

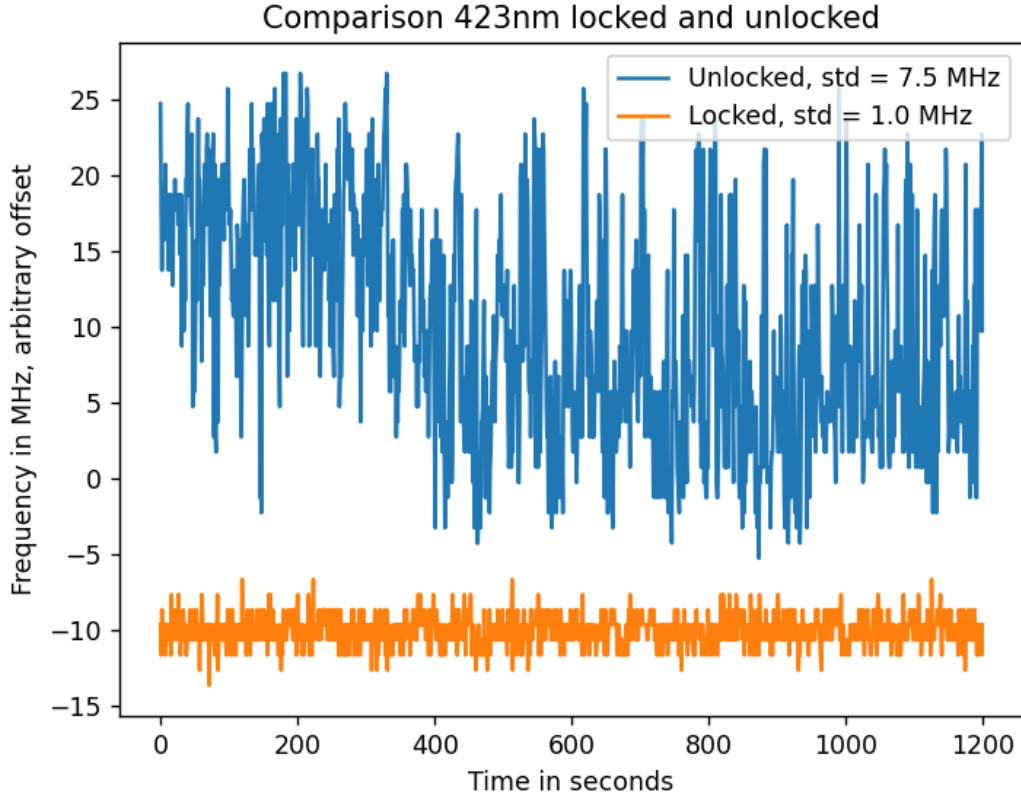


Figure 22: Variation of the frequency of the 423 nm laser over 20 minutes with the free-running laser (blue) and with the laser locked with polarization spectroscopy (orange). Arbitrary offsets are added for the two curves. Standard deviations are calculated over the whole 20 minutes. The lock-in amplifier parameters are: time constant $300 \mu\text{s}$, sensitivity 2 mV and a phase of 115° .

where the blue line corresponds to the free-running laser and the orange line to the laser locked with the polarization spectroscopy technique. The parameters leading to the locking situation in Figure 22 are listed below:

- The probe and pump beam have effective radii of 0.5 mm.
- The pump beam power is 6.6 mW.²⁴
- The probe beam power is 0.2 mW.
- The current in the optogalvanic lamp is 6 mA.

²⁴As it is chopped, the powermeter measures a 3.3 mW power.

The parameters for the lock-in amplifier are the same as in the previous subsection **4.1.1 Polarization spectroscopy signal** and are recall in the caption of Figure 22.

First, note that we subtract an arbitrary frequency offset for the two measurements to better view them in MHz unit. Initially, their average values are about 709 THz and the two signals are separated by several tens of MHz. Indeed, even if we tune the laser to a frequency close to that of the transition, the accuracy is limited to a few tens of MHz, because the wavemeter only displays digits up to the 100 MHz. The advantage of polarization spectroscopy is that it guarantees that the laser frequency is locked with respect to that of the $4^1S_0 \leftrightarrow 4^1P_1$ neutral calcium transition.

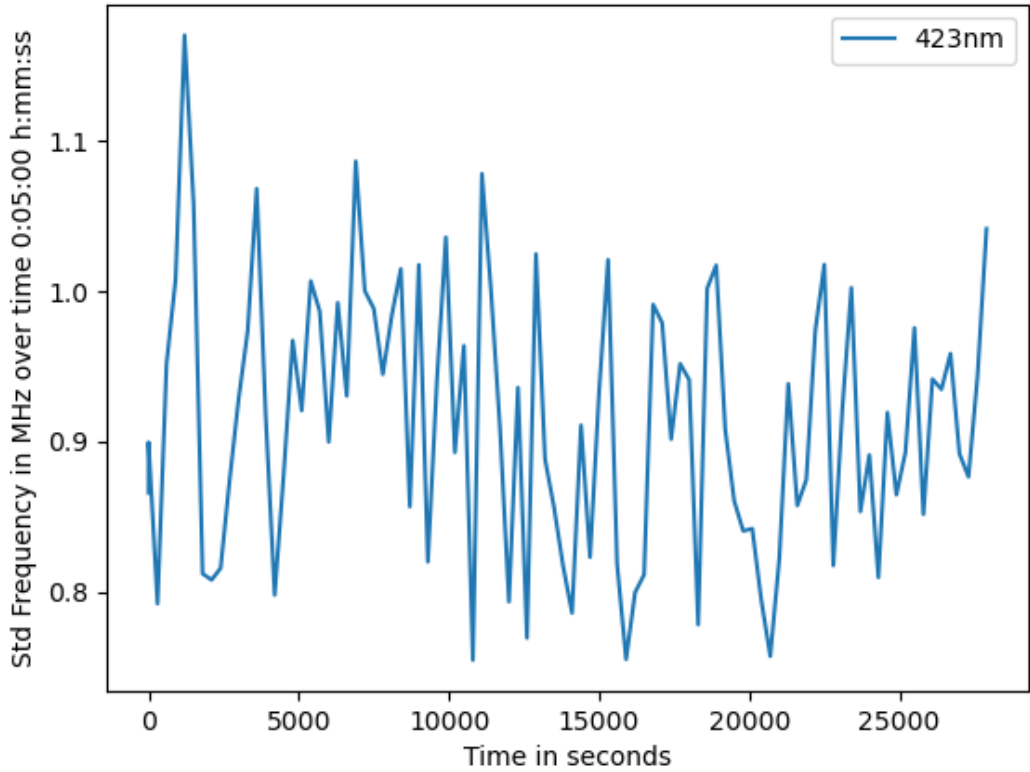


Figure 23: Standard deviation of the frequency calculated over 5 minutes of the 423 nm laser locked with polarization spectroscopy for 8 hours.

Secondly, we clearly see in Figure 22, that locking the 423 nm laser with the polarization spectroscopy improves its stability on short time scale of tens of seconds and on long time scale of 20 minutes. Thus, the polarization spectroscopy setup allows us to mitigate some technical noise on both short and long term, as discussed in the introduction subsection **1.2.1 Frequency**

noise. To further characterize the stability of the 423 nm locked laser, we measure the standard deviation of the frequency over 5 minutes for 8 hours, see Figure 23. This measurement shows that the stability of the laser is generally below 1 MHz. To measure the frequency of the laser, we use the wavemeter *WS6 - 200 Series* from *High Finesse* in the B25 laboratory. As mentioned in the subsection **3.3 HighFinesse WS6 - 200 Series wavemeter**, the measurement noise induced by this model of wavemeter is estimated to be about 0.6 MHz for the 729 nm light that we use as a reference for the other wavemeter in the B20 laboratory, so that some hundreds of kHz of the noise in Figure 23 is probably due to the wavemeter itself. However, the actual influence of the wavemeter on the 423 nm laser frequency measurement is not clear. Finally, according to the simulation, a stability of less than 1 MHz for the 423 nm laser should be sufficient to realize the two-photon cooling scheme for neutral calcium atoms.

4.2 Absorption drop in a three-level system

In this section, we present the results of the absorption drop in the three-level ladder system of neutral calcium atoms. Direct fitting of the data with the analytical expressions developed in the theoretical part leads to absurd parameters, such as temperatures of 1500 K for the calcium atomic vapor when a 7 mA current intensity is applied to the optogalvanic lamp. As a consequence, we perform additional measurements to fix some parameters. Therefore, before showing the results of the data fitting, we determine the optical densities and temperatures depending on the current intensities applied to the optogalvanic lamp.

4.2.1 Data parameters

The data displaying absorption drop is collected on an oscilloscope, model *TBS 1104* from *Tektronix*. The 733 nm Ti:Sapphire *M SQUARED SolsTiS* laser locked with the *étalon* is tuned to the resonance of the transition between the state 4^1P_1 and the state 4^1D_2 of neutral calcium atom. The 423 nm *Toptica DL pro* laser frequency is scanned by applying a voltage ramp to its piezoelectric controller. As in the case of polarization spectroscopy, we calibrate the voltage provided by the *EVIL* PI-controller to the laser frequency measured with the wavemeter to be able to transform the time axis into a frequency axis.

We repeat the measurements for different current intensities applied to the optogalvanic lamp, for different values of the 733 nm control beam power and

always keeping the same intensity for the 423 nm control beam. The values are shown in Table 4.

Intensity in mA		7	8	9	10	11
733 nm power in mW	Mean value	0	2.51	4.62	7.56	10.54
	Std	0	0.04	0.23	0.14	0.24
423 nm power ²⁵ in mW	Mean value	4.04				
	Std	0.04				

Table 4: Parameters to get transparency peaks within the absorption curves. The current intensities are those applied to the optogalvanic lamp. The powers values for the 733 nm and 423 nm beams are the ones measured before passing through the lamp.

In total, we have 25 curves because for each current intensity in Table 4, we collect data for all five values of the control beam power. Compared to polarization spectroscopy, we use a high-voltage power supply *Model PS310* from *Stanford Research Systems* so that the current intensity value is more precisely defined. Regarding the power of the probe and control beams, even though we try to keep the same values for the different experiments, they always fluctuate over time. For each experiment, we measure the power of the control beam and each time we change the current intensity, we measure the power of the probe beam. In total, we obtain five measurements for each power parameter whose mean value and standard deviation are displayed in Table 4.

4.2.2 Determining the optical densities

To facilitate the fitting of the data with the theoretical expression developed in section **2.5 Absorption drop in a three-level system**, we try to determine and fix some parameters. The first parameter we examine is the optical density

$$OD = -\log\left(\frac{I_{out}}{I_{in}}\right). \quad (109)$$

I_{out} is the output intensity and I_{in} is the input intensity of the probe beam as it passes through the atomic medium. To calculate the optical density due to the neutral calcium vapor in the optogalvanic lamp and not to take into account the intensity reflection coefficients of the windows of the lamp

²⁵When we measure the power of the probe beam with the powermeter we obtain half of this quantity, but we have to remember that we are chopping the signal so that half of the time the beam is blocked.

(108), we replace the input intensity I_{in} by the transmitted intensity when the lamp is off.

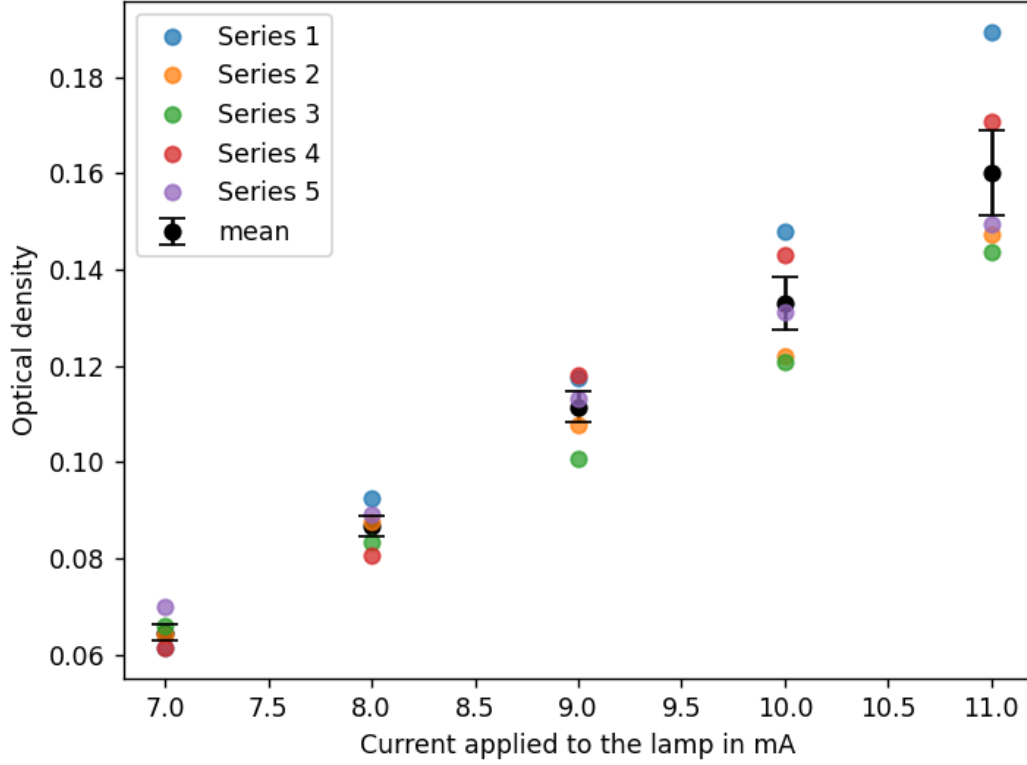


Figure 24: Optical density as a function of the current intensity applied to the optogalvanic lamp. The five series of measurement are displayed as well as the mean values (dark points) and the standard deviation (length of the error bar). These values correspond to the ones reported in Table 5.

The measurement of the optical density (109) is done with the 423 nm *Toptica DL pro* laser locked with the polarization spectroscopy setup. Then, the beam passes through the optogalvanic lamp with a current intensity set with a high voltage power supply *Model PS310* from *Stanford Research Systems*. For a fixed transmitted power of the probe beam of roughly $100 \mu\text{W}$ ²⁶ when the lamp is off, we decrease the current intensity applied to the lamp from 11 mA to 7 mA in 1 mA steps before turning the lamp off. The optical density values are then calculated using the expression (109) and we repeat the process five times and display it on Figure 24. The mean value and standard deviation of the optical density for each current intensity are reported

²⁶The probe beam is chopped, the corresponding measure of the powermeter is then $50 \mu\text{W}$.

in Table 5

Optical Density	Current intensity				
	7 mA	8 mA	9 mA	10 mA	11 mA
Mean Value	0.065	0.087	0.111	0.13	0.16
Standard deviation	0.003	0.004	0.007	0.01	0.02

Table 5: Mean value and standard deviation of the optical density as a function of the current intensity applied to the optogalvanic lamp. These values are displayed on Figure 24.

From now on, we will use the mean values in Table 5.

Note that we cannot directly use the optical density value (109) reported in Table 5. Indeed, the optical density depends on the parameters of the experiment as we can see by detailing the expression (109) with the general expression (43) of the linear complex dielectric susceptibility of the atomic medium

$$OD = \frac{\omega}{c} L \text{Im}(\chi^{exp}) = \frac{\omega}{c} L \frac{N}{V} \frac{|d_{12}|^2}{\hbar \epsilon_0} \frac{\text{Im}(\rho_{21}^{exp})}{\Omega^{exp}} = \alpha \frac{\text{Im}(\rho_{21}^{exp})}{\Omega^{exp}}. \quad (110)$$

The last term of the optical density expression (110) depends on the experiment we are performing, for example whether it is a two-level system or a three-level system. These particularities of the experiment will lead to a different coherent term ρ_{21}^{exp} and/or Rabi frequency Ω^{exp} in the expression (110). Nonetheless, the coefficient α defined in the expression (110) can be considered as depending only on the current intensity applied to the optogalvanic lamp. Indeed, the current intensity determines the atomic density of neutral calcium atoms. Then, even if we sweep the 423 nm laser frequency over a typical range of 1 GHz, this is negligible compared to its absolute frequency of 709 THz, so that its frequency is taken as a constant. Thus, if we determine the coefficient α (110) for each current intensity through the optical density measurements reported in Table 5, we could also use these values in the three-level configuration when adding the 733 nm laser.

Let's look at the detailed expression of the coefficient α (110) in the configuration we use to measure the optical densities in Table 5 using the steady state expressions for a two-level system (30)

$$\alpha = \frac{1}{OD} \cdot \frac{1}{\int w(v) dv \frac{\gamma_{12}}{2} \frac{1}{\delta(v)^2 + \gamma_{12}^2 + \Omega^2 \frac{\gamma_{12}}{\Gamma_{12}}}} \approx \frac{1}{OD} \cdot \frac{1}{\int w(v) dv \frac{\gamma_{12}}{2} \frac{1}{\delta(v)^2 + \gamma_{12}^2}} \quad (111)$$

$w(v)$ is the Maxwell-Boltzmann velocity distribution (46), $\delta(v)$ is the detuning with the Doppler shift (44). Looking at the expression of the integral (49), the approximation in expression (111) amounts to replacing the saturated linewidth of the transition γ_s (35) with the unsaturated linewidth of the transition γ_{12} . This is justified because the Rabi frequency, for a power of 100 μ W and a beam radius of 1 mm is about 14 MHz, is sufficiently low that we also get a low relative error

$$\frac{\gamma_s - \gamma_{12}}{\gamma_s} = 0.08. \quad (112)$$

Finally, to determine the coefficient α (111), it is sufficient to determine the temperature as a function of the current intensity such that we know the velocity distribution in equation (111). Once we have obtained α , the general form of the transmitted light intensity at the output of the optogalvanic lamp would only require to use the steady state expression for the coherent term, which we do for the three-level ladder system in **E.2 Approximated analytical expression**, and the Rabi frequencies for the two beams, as the absorption signal is

$$I_{out} = I_{in} \exp \left(-\alpha \frac{\text{Im}(\rho_{21}^{exp})}{\Omega^{exp}} \right). \quad (113)$$

4.2.3 Determining the temperatures

To determine the temperature of the atomic vapor inside the optogalvanic lamp, we use the data set for different current intensities and with the 733 nm laser turned off, see Table 4. The corresponding curves are the colored lines plotted in Figure 25 and arbitrarily shifted by 1 GHz to better distinguish them. At first, we see in Figure 25 that we lack information because we only have the tip of the absorption peak and not the tails of the signal converging to the value when the light is too far detuned to interact with the atoms. Indeed, the frequency range we can scan with the *Toptica DL pro* laser is limited to a few hundreds of MHz because of mode hops. However, we can recover enough information to determine temperature by using the five curves together. Indeed, the probe beam is the same in all five experiments, so it follows that the input intensities and the Rabi frequencies are the same. The differences between the five curves are:

- The calcium atomic density, which depends on the current intensity applied to the optogalvanic lamp.
- The temperature, which changes the expression of the coherent terms in the output signal (113) due to the Doppler broadening.

Though, as we know the optical densities in Table 5, the atomic densities can be viewed as a function of temperatures.

Therefore, by introducing one parameter for the input intensity for all five curves and a temperature parameter for each curve, we can fit all the curves at once. To do this, we use the steady state expression for the coherent term for a two-level system (30), the expression of the coefficient α (111), along with the optical density values set in Table 5. The result of the fit are represented by the black dashed lines in Figure 25.

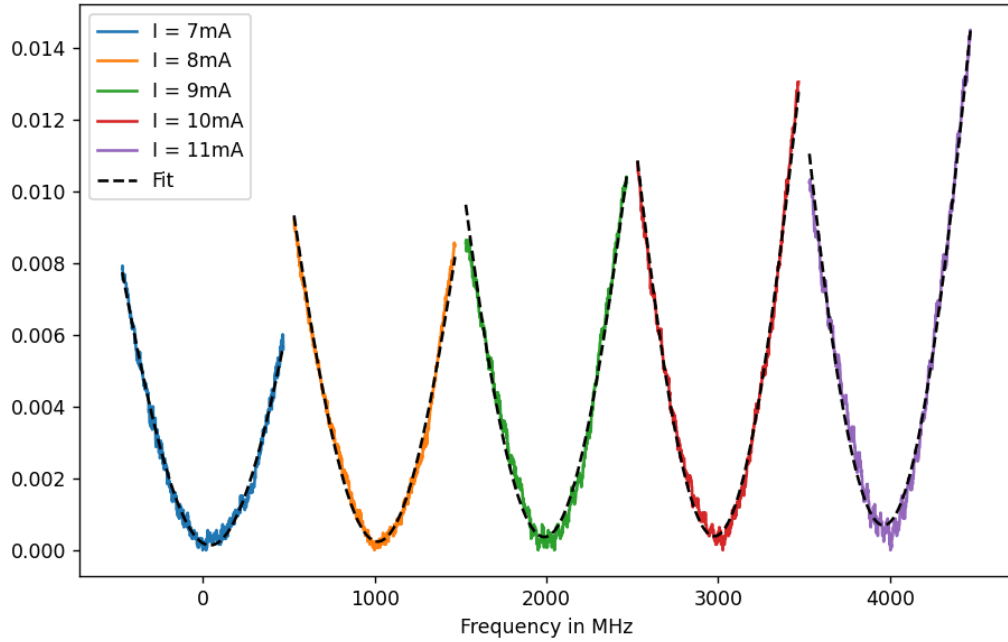


Figure 25: Absorption curves with the 733 nm beam turned off (colored) and the fitted curves (black dashed), for a single amplitude fitting parameter and individual temperature parameters. For each current the curves are arbitrarily displaced by 1 GHz. The fitted temperatures are reported in Table 6 and plotted in Figure 26.

In the fitting function, we fixed the 423 nm Rabi frequency to 60 MHz which is the typical value we expect for the probe beam power equal to 4.04 mW, see Table 4, and a radius of about 1 mm. The curve fit is shown in Figure 25. The fitted temperature parameters are reported in Table 6 and displayed in Figure 26.

A first remark concerns the fixed value of the Rabi frequency. The value of 60 MHz is a rough estimate so we verify that changing it by ± 20 MHz, only changes the fitted temperature parameters by 3 – 4 K, which is smaller than the standard deviation we obtain for the fitted temperatures, see Table 6.

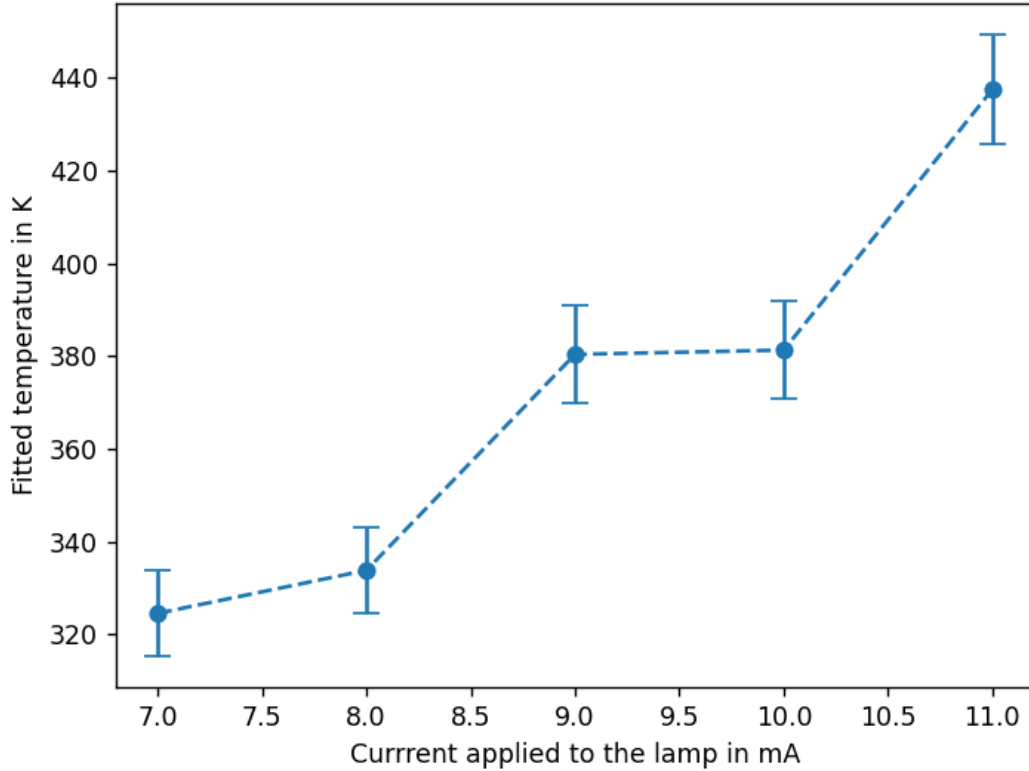


Figure 26: Fitted temperatures as a function of the current intensity applied to the optogalvanic lamp. The error bars correspond to the standard deviations reported in Table 6.

For this reason, we retain the temperature values reported in Table 6 to fit the data in the next subsection.

The second remark is that the temperature in the 9 mA current case seems to be high compare to the other cases, see Figure 26. One possible explanation is that since this was the very first measurement of the 25 ones, it might be that the thermal equilibrium state inside the optogalvanic lamp was not reached.²⁷

A third remark is that it is important to fit all five curves at the same time with only one fitting parameter for the amplitude because we only get a limited part of the gaussian-like absorption signal, see Figure 25. Because of this limitation, we only get the center of the signal and not the tail end of the gaussian. Fitting the curves individually, so having for each curve one parameter for the amplitude along with the five temperature parameters,

²⁷The information we found on hollow cathode lamp is that the typical heating time is about 10 – 15 minutes [31] and could change depending on the atomic species.

Temperature	Current intensity				
	7 mA	8 mA	9 mA	10 mA	11 mA
Mean Value	323	334	383	385	444
Standard deviation	19	19	21	21	24

Table 6: Mean value and standard deviation of the temperatures for each current intensity applied to the optogalvanic lamp. These values corresponds to the ones we get from the fits on Figure 25 and are displayed on Figure 26.

we obtain absurd values for temperature. It ranges from 197 K for a 9 mA current to 1583 K for a 7 mA current. However, we know that a higher current is equivalent to a higher temperature inside the optogalvanic lamp. Moreover, we can compare the results with the temperatures for currents of 30 mA to 70 mA in a hollow cathode lamp for neutral calcium atoms which are of the order of 750 K to 1200 K [33].

With the temperature values from Table 6 and the optical density values from Table 5, we are able to calculate the coefficients α (111). In conclusion, the only remaining parameters we need to consider for fitting the data, are those that determine the ratio of the steady state coherent term ρ_{12} to the Rabi frequency of the probe beam, for which we get a theoretical expression developed in **Appendix E Three-Level Ladder System**.

4.2.4 Fitting the data

Now that we have the values of the optical densities in Table 5, the temperatures in Table 6, and that we can easily calculate the coefficient α (111), the parameters that remain free and that we must adjust are:

- The effective radius of the 423 nm laser. As the value of the probe beam power in Table 4 and the intensity reflection coefficient (108) are fixed, the radius allows to adjust the Rabi frequency of the 423 nm laser. This parameter is the same for all 25 measurements.
- The effective radius of the 733 nm laser, for analogous reasons to those of the effective radius of the 423 nm laser. This parameter is also unique for the 25 measurements.
- The detuning of the control 733 nm laser, which is different from one measurement to the other as we only use the *étalon* locking of the Ti:Sapphire and not the reference cavity which is more stable.

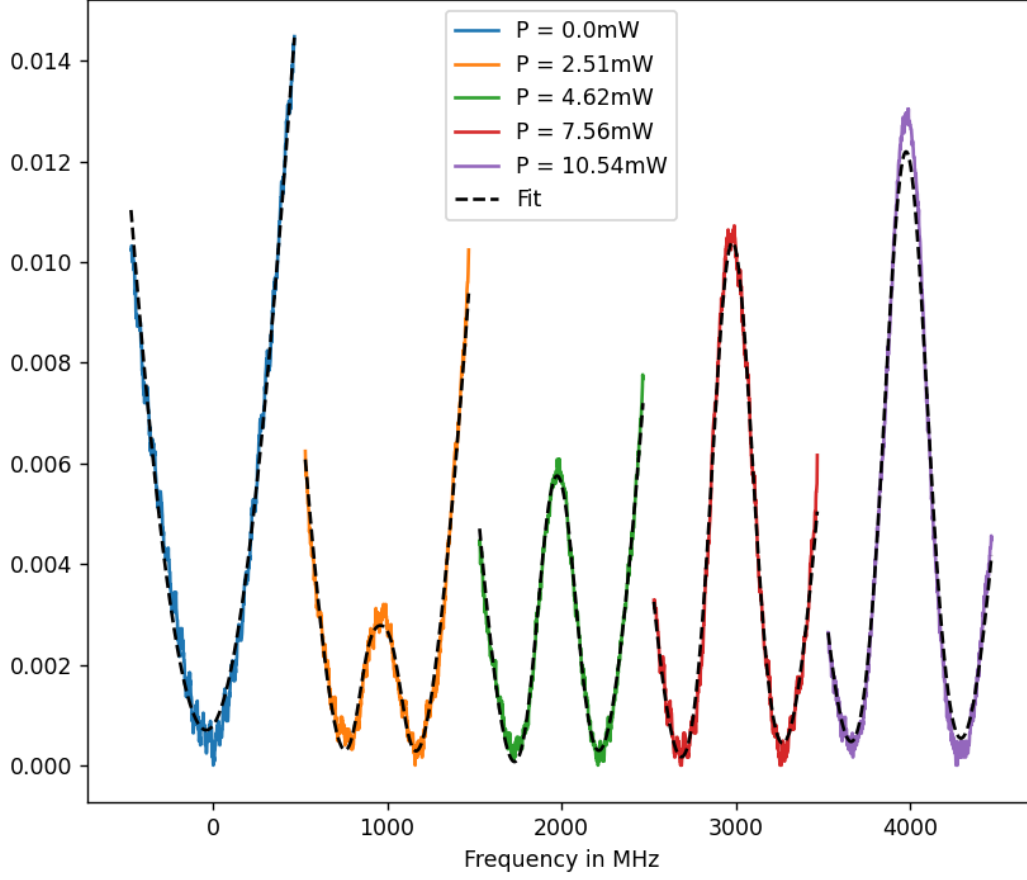


Figure 27: The absorption drop curves (colored) in the three-level ladder system of neutral calcium for a current intensity of 11 mA applied to the lamp are arbitrarily displaced by 1 GHz. Some of the parameters used to plot the fitted curves (black dashed) are reported in Table 7.

- Likewise, we introduce a frequency shift for the 423 nm probe laser for each measurement.
- The voltage amplitude of the signal that multiplies the exponential absorption part of the signal (113). This parameter is the same for all measurements because the intensity of the probe beam is fixed.
- The empirical error probabilities (96) that we introduce in **2.5.6 Modelling influence of collisions**. We introduce such a probability for each measurement.
- Eventually, for each curve, we introduce a general offset that should be close to zero.

To fit the data we use the expression for the coherent term of the three-level ladder system developed in **E.2 Approximated analytical expression**. Although we fit the 25 data curves (colored) at the same time due to the shared parameters, we have displayed the fitted curves (black dashed) only for the set corresponding to the 11 mA current intensity in Figure 27.²⁸ In this figure, the curves are arbitrarily shifted by 1 GHz to better distinguish them. Some of the fitted parameters are tabulated in Table 7.

Current intensity = 11 mA		733 nm power in mW				
		0	2.51	4.62	7.56	10.54
Radius 423 nm	Mean	0.76 mm				
	Std	10^{-6} mm				
Radius 733 nm	Mean	0.76 mm				
	Std	10^{-6} mm				
Amplitude	Mean	0.866 V				
	Std	0.009 V				
Error probability (1 - p)	Mean	0.405	0.601	0.887	1.00	
	Std	0.002	0.002	0.002	0.002	

Table 7: Fitted parameters for the case of a 11 mA current intensity applied to the optogalvanic lamp, though they are found by fitted the 25 measurements. The corresponding curves are plotted in Figure 27.

The fitted radii, in Table 7, are of the right order of magnitude, although they are smaller than the measured value of 0.85 mm that we obtain with the beam profiler for the 423 nm beam and the 733 nm beam. However, we realigned the experiment after taking the measurements plotted in Figure 27 and before measuring the beams radii with the beam profiler, because strong reflection effects can occur with the lamp, it is possible that the beams were distorted which would modify their fitted radii. Another possible explanation is that we have measured the optogalvanic lamp window intensity reflection coefficient (108) for the 423 nm laser. As a consequence, the reflection coefficient in intensity for the 733 nm laser could be different which would influence the radius of the 733 nm to get the right Rabi frequencies. Also note that in all the previous calculations and models, we consider uniform beams and therefore uniform Rabi frequencies. This is to keep things simple. Especially, since the beams are not perfectly gaussian but have a more complex shape that would have been difficult and time-consuming to integrate the expression of the steady state coherent term as a function of the beams shapes and the relative positions of their centers.

²⁸We perform the fit with the `curve_fit` function from the `scipy.optimize` library.

The empirical error probabilities ($1 - p$) are reported in Table 7. If we had more time, we could have looked for an empirical model for these probabilities and explained them as a function of some physical parameters of the atomic vapor inside the lamp. In addition, when performing our fit, we only consider the atomic density instead of the difference of atomic densities between the ground state and the excited state. By quickly examining the consequences of this approximation, we find that most of parameters have not been changed significantly. The relative differences are generally small, for instance temperatures are changed by 1 – 4 K, except in particular for these empirical probabilities, though they are still necessary to fit the data. Therefore, if one wishes to examine the physical explanation behind these empirical probabilities, it would be important to consider the difference of atomic densities instead of just the calcium atomic density.

4.3 423 nm - 733 nm Two-color polarization spectroscopy

In this section, we present the results obtained for the two-color polarization spectroscopy: the dispersion-like signal and the 733 nm laser frequency stability when it is locked with this spectroscopic technique.

4.3.1 Two-color polarization spectroscopy signal

To obtain the two-color polarization spectroscopy signal, we lock the 423 nm laser with the polarization spectroscopy technique and scan the Ti:Sapphire *M SQUARED SolsTiS* laser around the 733 nm wavelength. The dispersion-like signals in Figure 28 are recorded on an oscilloscope, model *TBS 1104* from *Tektronix*. Previously, in the polarization spectroscopy and to see transparency peaks in the absorption curves, we scanned the 423 nm *Toptica DL pro* laser frequency and we had to calibrate the voltage provided by the *EVIL* PI-controller to the laser frequency using the wavemeter. Since the internal controller of the CW Ti:Sapphire *M SQUARED SolsTiS* laser allows us to set the swept frequency range directly, we do not need to do this calibration here. The time axis is converted to a frequency axis by finding the coordinates of the extrema of the voltage ramp, which is applied to the piezoelectric controller of the laser and is displayed on another channel of the oscilloscope, the difference of which corresponds to the frequency range.

We collect the data with the experimental setup shown in Figure 11 for the fixed parameters:

- The current intensity applied to the optogalvanic lamp is 7 mA.

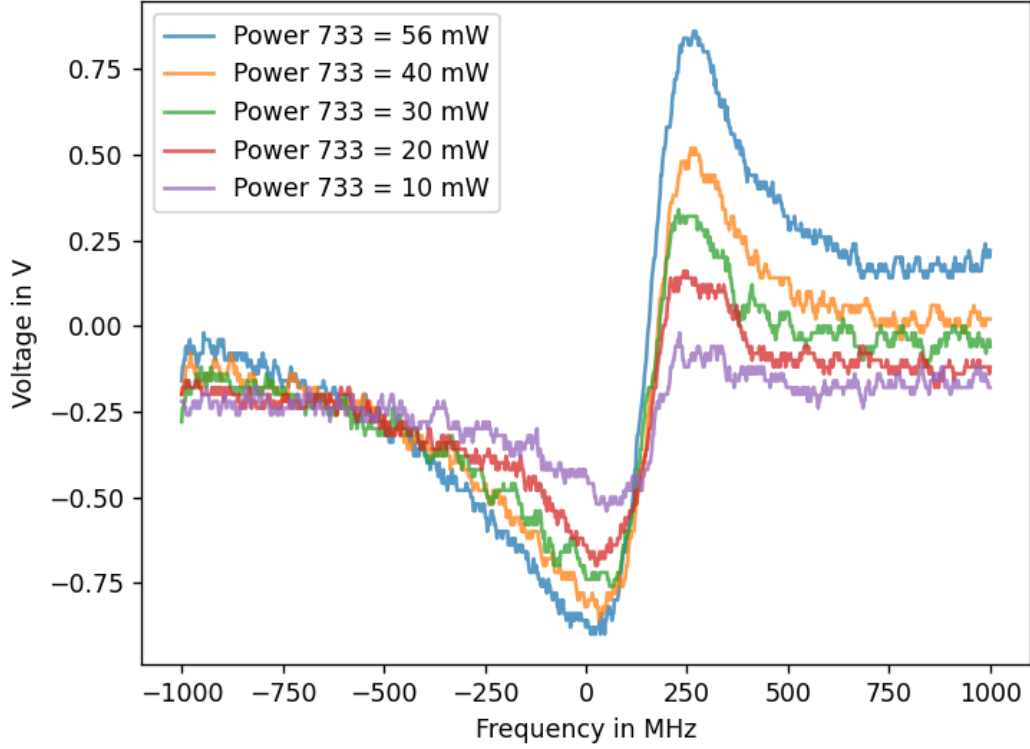


Figure 28: Two-color polarization spectroscopy dispersion-like signal. Power of the 423 nm: 4.4 mW. Current intensity applied to the optogalvanic lamp: 7 mA. The lock-in amplifier parameters are: time constant 1 ms, sensitivity 20 mV and a phase of -50 deg.²⁹

- The probe beam power is 4.4 mW.
- The radii of the probe and the control beam are approximately the same and evaluated to 0.85 mm, according to the gaussian fit realized with the beam profiler from *Thorlabs*.
- The time constant of the lock-in amplifier is 1 ms.
- The sensitivity of the lock-in amplifier is 20 mV.
- The swept frequency range for the 733 nm control beam is 2 GHz.

²⁹Fitting the data in Figure 28 with the model presented in section **2.6 Two-color polarization spectroscopy** and plotted in Figure 13 (b) is out of question, as a single simulation already takes a few tens of minutes.

To obtain the different curves in Figure 11, we only change the control beam power, the values are $\{10, 20, 30, 40, 56\}$ in mW.³⁰

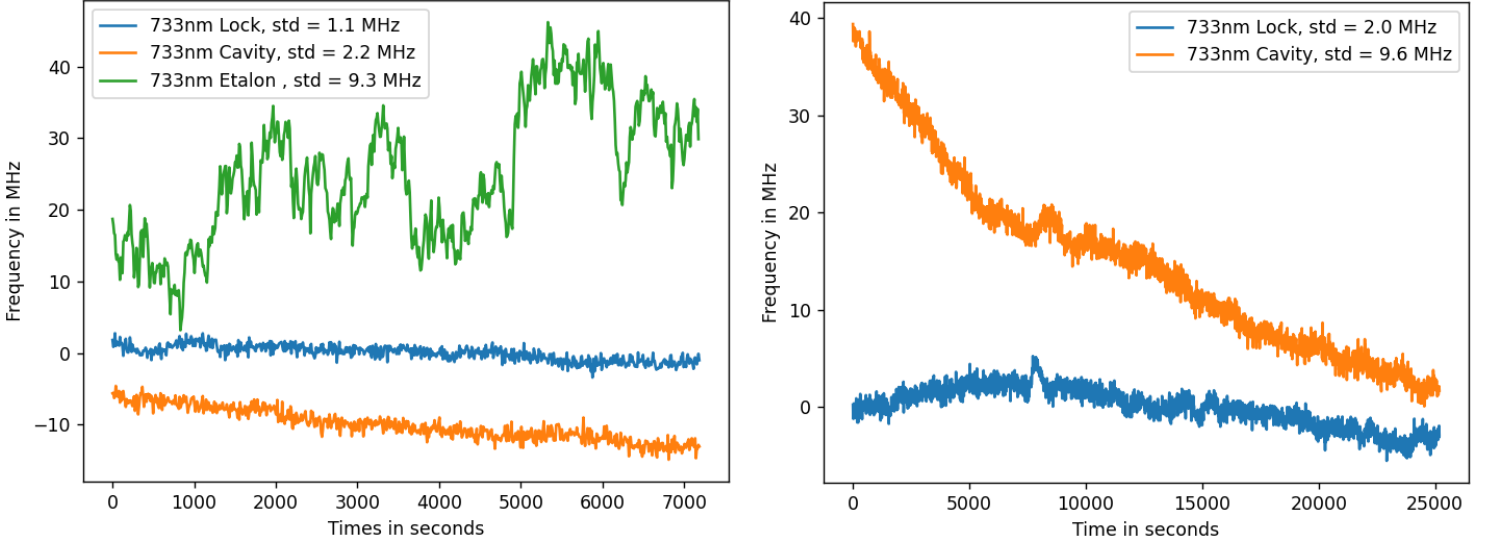
In Figure 28, we can see the effect of changing the 733 nm control beam power while keeping all other parameters constant. Changing the control power does not significantly modify the width of the dispersion-like signal, as this characteristic is essentially set by the Doppler width. However, to some extent, the higher the control beam power, the higher the amplitude of the resulting signal. Therefore, the slope around zero can increase with higher 733 nm laser power, resulting in better frequency stability when using this signal to lock the *SolsTiS* laser. As shown in Figure 28, the slope corresponding to the highest power we can achieve is also the steepest, so we work with the highest power possible. One possibility that could be explored to achieve a steeper slope would be to increase the current intensity applied to the optogalvanic lamp. This would increase the temperature and thus the width of the signal, but in the meantime it would also increase the atomic density of the calcium neutral atoms and the amplitude of the signal, which could provide a better signal to lock the 733 nm laser.

4.3.2 Locking of the 733 nm laser

Now that we get the dispersion-like signal as in Figure 28, we can lock the 733 nm laser on the neutral calcium transition between the state 4^1P_1 and the state 4^1D_2 . As with the polarization spectroscopy, the output signal from the lock-in amplifier is sent to the *EVIL* PI-controller [40] and we use the *DevilClient* software to set the proportional and integral parameters of the control loop. The corrected voltage is sent back to the *SolsTiS* control unit also called *ICE-Bloc*. As detailed in the manual of the *SolsTiS* laser [12] and the note on locking the laser to a reference cavity [41], we feedback using the slow control channel of the piezo-mounted mirror. The main reason for this choice is the control ranges and sensitivities of the fast and slow external inputs. For the slow (fast) external input, the control range is 30 GHz (80 MHz) and the sensitivity is of 1.5 GHz/V (4 MHz/V). First, as we can see in Figure 28, the frequency scale of the dispersion-like signal is typically 1 GHz and in addition, since the *EVIL* PI-controller allows only a few volts and not 20 V to be swept, the frequency scan range of the fast external input is not relevant.

The resulting locking of the 733 nm laser is shown in Figures 29 (a) and (b). The parameters use to lock the laser are:

³⁰As the control beam is chopped the measured powers with the powermeter are $\{5, 10, 15, 20, 28\}$ in mW



(a) Frequency of the 733 nm stabilized over 2 hours with the two-color polarization spectroscopy (blue), the reference cavity (orange), and the *étalon* (green). (b) Comparison of the stability over 7 hours of the 733 nm with the two-color polarization spectroscopy technique and with the reference cavity of the *SolsTiS* laser.

Figure 29: Comparison of the frequency stability of the 733 nm laser for the different stabilization options: the two-color polarization spectroscopy, the reference cavity, and the *étalon*. For the two-color polarization spectroscopy, the parameters are: the 423 nm power is 4.3mW, the 733 nm power is 80 mW, for the lock-in amplifier the time constant is 1 ms, and the sensitivity is 2 mV.

- The intensity in the optogalvanic lamp is 7 mA.
- The 423 nm probe beam power is 4.3 mW.
- The 733 nm control beam power is 80 mW. With the powermeter we measure 40 mW as the beam is chopped.
- The effective radii of the two beams are 0.85 mm.
- The time constant of the lock-in amplifier is 1 ms.
- The sensitivity of the lock-in amplifier is 2 mV.

As in the case of the polarization spectroscopy for the 423 nm laser, we arbitrarily subtract a frequency offset from the different signals, because with two-color polarization spectroscopy, the 733 nm laser frequency is stabilized with respect to the transition between the excited states 4^1P_1 and 4^1D_2 of

the neutral calcium atoms, but with the internal stabilization options of the *SolsTiS* laser, we rely on the precision of the *WS6 - 200 Series* wavemeter.

In Figure 29 (a), we compare the frequency stability of the 733 nm laser with the different options available to us: two-color polarization spectroscopy (blue), the *SolsTiS* reference cavity (orange), and the *SolsTiS étalon* (green). We find that the frequency stability using only the *étalon* is worse than for the other two options, which is logical as we use both in addition to the locking of the *étalon*. Therefore, we compare the stability with the reference cavity and with the two-color polarization spectroscopy over a longer time scale of 7 hours in Figure 29 (b). On this longer time scale, we can clearly see that the long-term drift of the two-color polarization spectroscopy technique is smaller than that experienced by the reference cavity of the *SolsTiS* laser. However, on short time scale of a few tens of seconds, the stability seems to be equivalent.

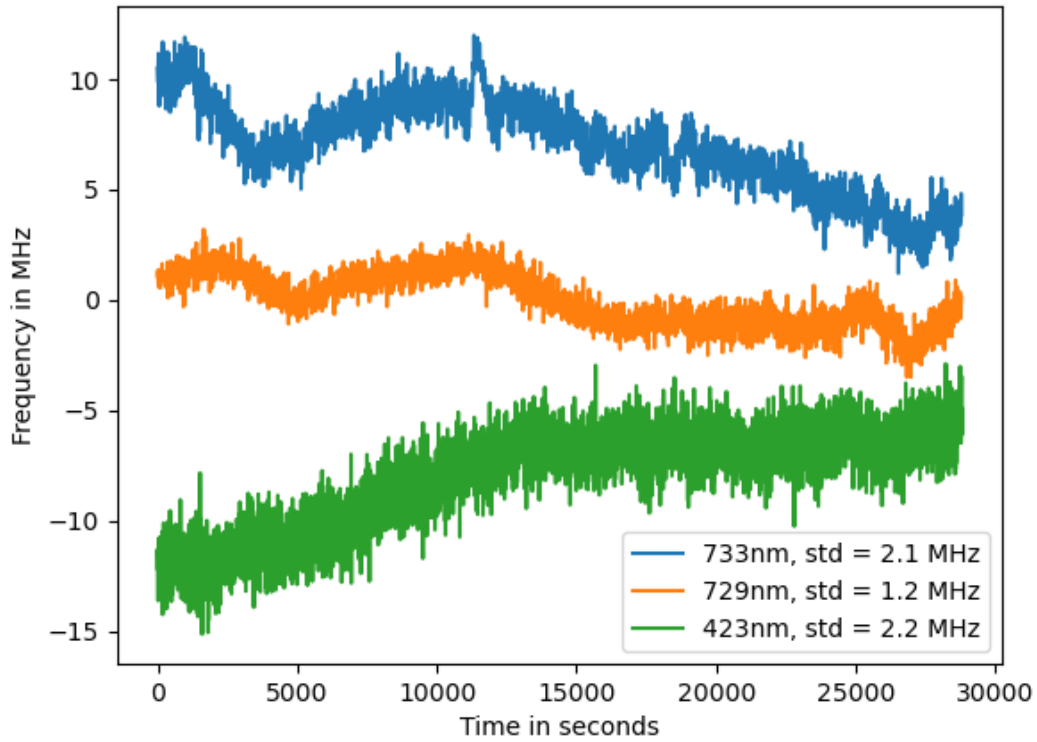


Figure 30: Frequencies of the 733 nm and the 423 nm lasers locked respectively with two-color polarization spectroscopy and polarization spectroscopy over 8 hours. The 729 nm is locked to a high finesse cavity and is used as a reference for the wavemeter which is used to read the 733 nm frequency.

In an attempt to understand the remaining instability of the 733 nm

laser when locked with the two-color polarization spectroscopy, we examine the frequency correlations between the 733 nm, the 729 nm, and the 423 nm lasers that we recorded simultaneously for 8 hours, see Figure 30.

We expect to observe long term correlations between the 733 nm and the 423 nm frequencies, as they essentially verify the two-photon resonance condition. The correlations between the 733 nm frequency and the 729 nm frequency should come from the fact that, since we consider the 729 nm as a reference because it is locked to a high-finesse cavity, the long term drift of the 729 nm frequency should be caused by the wavemeter with which we also measure the 733 nm frequency. However, in Figure 30, these correlations are not significant. For instance, during the last hours of the measurement, the

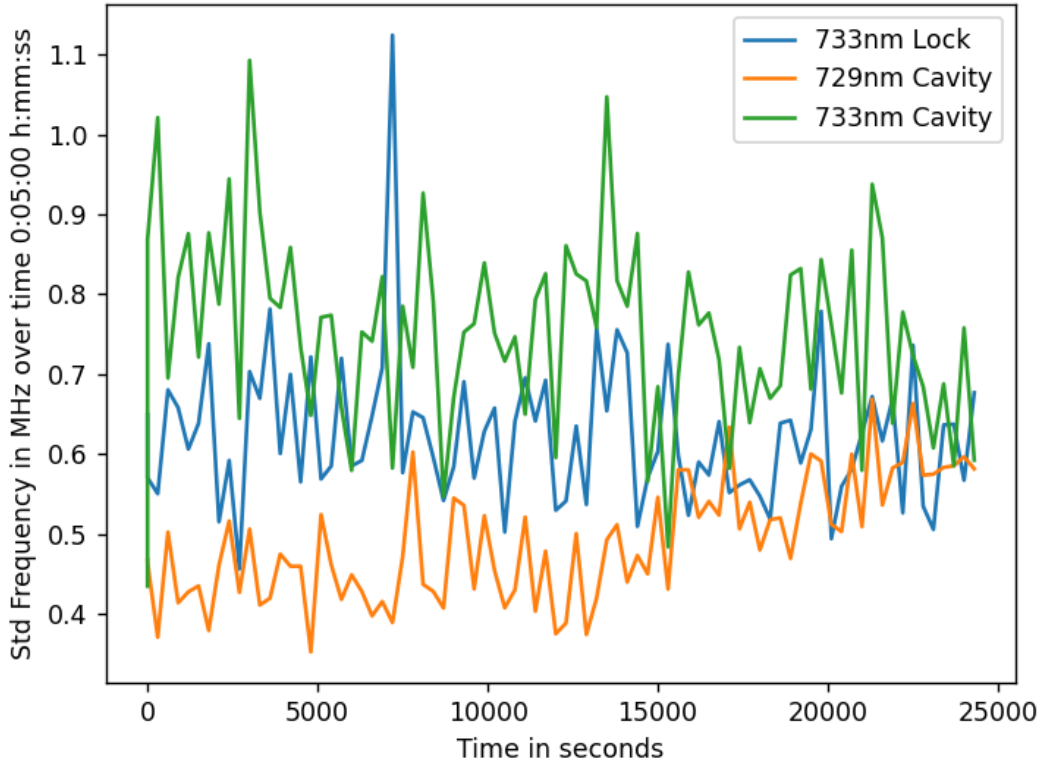


Figure 31: Comparison of the frequency standard deviation calculated over 5 minutes for 7 hours. The 733 nm is locked with the two-color polarization spectroscopy technique (blue) and with the reference cavity of the *SolsTiS* laser (green). The 729 nm laser (orange) is locked to a high finesse cavity and is used as a reference.

423 nm and 729 nm lasers frequencies are relatively constant, while the 733 nm frequency decreases. However, it should be remembered that we use two

different wavemeters in two different laboratories to record on the one hand the 423 nm laser frequency and on the other hand the 733 nm and 729 nm lasers frequencies. Thus, they could be subject to different and uncorrelated drifts.

Eventually, we can study the stability of the 733 nm frequency over a short time scale by examining the standard deviation calculated over 5 minutes for 7 hours, see Figure 31. First, we note that the standard deviation of the 733 nm laser locked with two-color polarization spectroscopy is slightly smaller than that which we get using the reference cavity and is generally less than 0.8 MHz.³¹ From the simulation, this would be stable enough for us to achieve the two-photon cooling scheme. The standard deviation of the frequency of the 729 nm reference laser in Figure 31 also gives us an idea of the noise induced by the finite precision of the wavemeter. Even though, in the latter part of the measurement most of the 733 nm frequency instability could be explained by the measurement noise of the wavemeter, we find that during the first few hours of the measurement the wavemeter alone cannot explain all of the 733 nm frequency instability.

4.4 Rydberg excitation

Similar to what we did with the three-level ladder system setup, we briefly examine the excitation to a low Rydberg state. Instead of using the 733 nm laser, we use a 403 nm *Toptica DL Pro* laser so that the third state of the ladder system is the state 12^1D_2 instead of the state 4^1D_2 of neutral calcium atom. Since the lifetime of the Rydberg state is generally much longer and that the two wavelengths are closer, which limits the Doppler broadening, it could be interesting to see if we can reach the regime of electromagnetically induced transparency, as it has been done with erbium [42].

In Figure 32 we check for the presence of the 12^1D_2 state near the frequency recorded in the NIST database of 743.4681 THz [43]. To do this, we lock the 423 nm with the polarization spectroscopy technique and we sweep the frequency of the 403 nm laser. On the contrary of the three-level ladder system setup, it is the 403 nm control laser beam that is chopped at 100 kHz via an arbitrary waveform generator, a switch and an acousto-optic modulator and not the 423 nm probe beam. The output signal is sent to a lock-in amplifier to filter out noise. A noteworthy difference from the previous se-

³¹One difference between the two data sets is that in the case of the reference cavity, we were using the *Sprout-G* pump laser from *Lighthouse Photonics* at its maximum output power of 18 W and in the case of two-color polarization spectroscopy at an output power of 16 W. Indeed, at 16 W the intensity seems to be more stable, we do not know to what extent this could affect the frequency stability of the Ti:Sapphire laser.

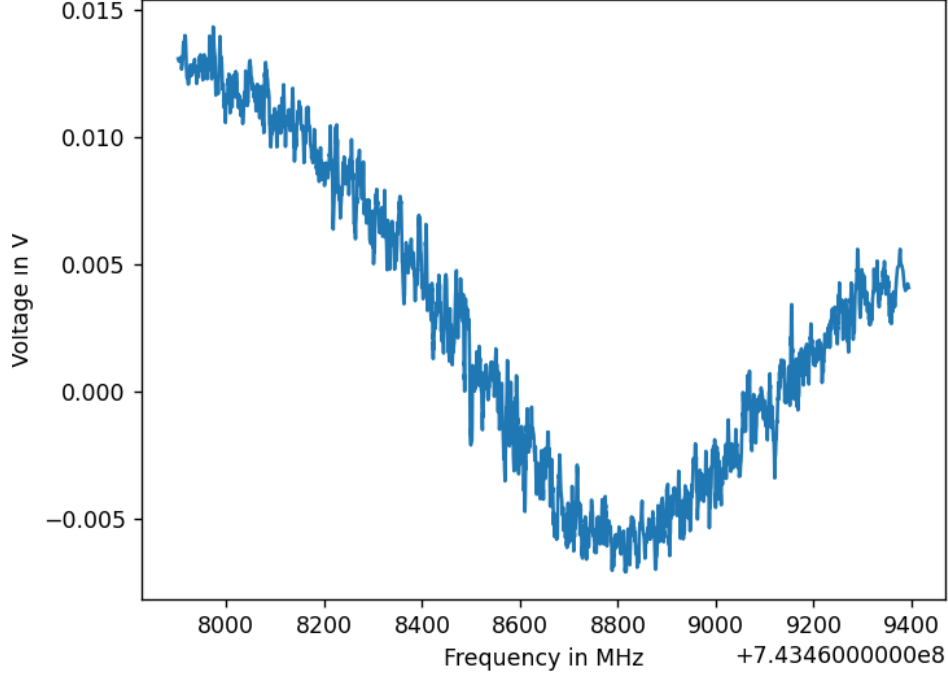


Figure 32: Absorption peak at the two-photon resonance in the three-level system of the neutral calcium atoms $\{4^1S_0, 4^1P_1, 12^1D_2\}$. Power of the 423 nm: 6.7 mW, radius of 0.5 mm. Power of the 403 nm: 22 mW, radius of 1.0 mm. Time constant of the lock-in amplifier: 3ms. Sensitivity of the lock-in amplifier: 200 mV

tups is that we use a telescope to decrease the size of the 423 nm beam by a factor of 3 and to correct for the expansion of the 423 nm beam. Thus, the effective beam diameter measured with the beam profiler from *Thorlabs* is 0.5 mm. Likewise, the effective beam diameter for the 403 nm laser beam is 1.0 mm. The telescope allows to obtain a higher Rabi frequency while using the same amount of power for the 423 nm beam and thus obtain a higher amplitude for the resulting signal. We quickly try to observe a signal without the telescope but we cannot see anything.

To get the Figure 32, which is the absorption peak due to the presence of the 12^1D_2 state as a function of the 403 nm laser frequency, we read the frequency on the wavemeter, the voltage on the oscilloscope and we take the rolling average on 10 points to get a smoother signal. The peak at 743.4688 THz corresponds to the absorption due to reaching the two-photon resonance

between the 4^1S_0 state and the 12^1D_2 state. Since we see this absorption peak, it might be possible to implement the same two-color polarization spectroscopy technique, which we use to stabilize the 733 nm laser on the $4^1P_1 \leftrightarrow 4^1D_2$ transition, to stabilize in this case the 403 nm laser on the $4^1P_1 \leftrightarrow 12^1D_2$ transition of neutral calcium atoms [44].

5 Conclusion

In this master's thesis we study the theory and we implement two frequency stabilizing techniques. We start with the polarization spectroscopy for the 423nm beam between the 4^1S_0 ground state and the 4^1P_1 excited state of neutral calcium atoms. The resulting frequency stability for this beam is of the order of 1 MHz. Then we continue with the two-color polarization spectroscopy with the additional 733 nm laser and the 4^1D_2 state. We also reach a frequency stability of about 1 MHz for the 733 nm Ti:Sapphire laser for the transition between the 4^1P_1 state and the 4^1D_2 state.

In addition, we develop an empirical model to fit the transparency peak we get in the Autler-Townes splitting regime as we are using a quite powerful 733 nm control beam. Indeed, the transparency peak tends to disappear faster than what we expect from exact calculation of the steady state solution of the optical Bloch equations. This observation forced us to introduce an empirical error probability to fit the collected data. We think that this might be due to collisions inside the lamp. Hence, as the simulations taking into account velocity distribution predict an even smaller peak in the electromagnetically induced transparency regime we doubt whether it would be possible to reach it in the present configuration.

Eventually, the main goal which underlies this master's thesis is to be able to use these frequency stabilized lasers to realize a two-photon cooling scheme. According to simulations, the 1 MHz frequency stability that we demonstrate from the measurements with the wavemeters for both lasers should be enough to realize this cooling technique. Nonetheless, the question remains whether we could effectively accomplish this sub-Doppler cooling or not. Besides, some elements are still unclear, notably the influence of the wavemeters on the frequency measurements and the determination of the parameters allowing to get an optimal stability in the two spectroscopic setups. Regarding the first problem, a beat note measurement of the lasers with reference lasers locked to high finesse cavities could be a solution to obtain a more accurate characterization of the frequency stability. For the second problem, it would be necessary to dive into more detailed theoretical models and simulations to get rid of the empirical parameters we introduced.

Appendix A Simple Case of Lindblad Operator

In this appendix we propose a toy model to derive the form of the Lindblad operator with its jumps operators (23). To do so, we make two strong assumptions about the evolution of the system due to its interactions with the environment.

First, as in [45] we assume that the environment is decoupled from the state of the system because it is a kind of a reservoir and is independent of time, or at least it evolves on a much longer time scale than the system of interest. Therefore, the density matrix operator for the system S and the environment E is of the form

$$\rho(t) = \rho_S(t) \otimes \rho_E(0). \quad (114)$$

The second assumption we introduce to obtain a simple model is that the completely positive and trace preserving map representing the evolution of the density operator during an infinitesimal time dt from t to $t + dt$ is independent of time t . So that in the operator-sum representation [15] the evolution of the system and the environment is

$$\rho(t + dt) = \mathcal{V}(\rho(t)) = \sum_l V_l \rho(t) V_l^\dagger. \quad (115)$$

The operators $\{V_l\}_l$ ³² verify the condition

$$\sum_l V_l^\dagger V_l = \mathbf{1}_{S \times E}. \quad (116)$$

The first condition (114) implies that the time derivative of the system density matrix is

$$\frac{\partial \rho_S}{\partial t} = \text{Tr}_E \left(\frac{\partial \rho}{\partial t} \right) = \text{Tr}_E \left(\frac{\rho(t + dt) - \rho(t)}{dt} \right). \quad (117)$$

Using the second (115) and third relations (116), we can express the right-hand side of the equation (117) as

$$dt \frac{\partial \rho_S}{\partial t} = \text{Tr}_E \left(\sum_l V_l \rho(t) V_l^\dagger - \frac{1}{2} \left(V_l^\dagger V_l \rho(t) + \rho(t) V_l^\dagger V_l \right) \right). \quad (118)$$

³²One example of such operators are $\{\sqrt{1-p} \mathbf{1}, \sqrt{p} X\}$, which we introduced in subsection **2.5.6 Modelling influence of collisions** and which describe a bit flip error.

We introduce an orthonormal basis $\{|n\rangle\}_n$ of the Hilbert space of the environment E , which diagonalizes the density operator $\rho_E(0)$

$$\rho_E(0) = \sum_n \lambda_n |n\rangle\langle n|, \text{ with } \lambda_n \geq 0. \quad (119)$$

We recall the useful mathematical relations

$$\begin{cases} \text{Tr}_E [A \cdot (B_S \otimes C_E)] &= \text{Tr}_E [A \cdot (\mathbf{1}_S \otimes C_E)] \cdot B_S \\ \text{Tr}_E [(B_S \otimes C_E) \cdot A] &= B_S \cdot \text{Tr}_E [(\mathbf{1}_S \otimes C_E) \cdot A] \end{cases} \quad (120)$$

Then developing the relation (118) we get

$$\begin{aligned} dt \frac{\partial \rho_S}{\partial t} &= \sum_{l,n} (\mathbf{1}_S \otimes \langle n|) \cdot V_l \rho(t) V_l^\dagger \cdot (\mathbf{1}_S \otimes |n\rangle) - \frac{1}{2} \sum_{l,m} \lambda_m \cdot \\ &\quad \text{Tr}_E \left[V_l^\dagger V_l (\rho_S(t) \otimes |m\rangle\langle m|) + (\rho_S(t) \otimes |m\rangle\langle m|) V_l^\dagger V_l \right] \\ &= \sum_{l,n,m} \lambda_m (\mathbf{1}_S \otimes \langle n|) \cdot V_l \cdot (\mathbf{1}_S \otimes |m\rangle) \rho_S(t) (\mathbf{1}_S \otimes \langle m|) \cdot \\ &\quad V_l^\dagger \cdot (\mathbf{1}_S \otimes |n\rangle) \\ &\quad - \frac{1}{2} \sum_{l,m} \lambda_m (\mathbf{1}_S \otimes \langle m|) V_l^\dagger \cdot (\mathbf{1}_S \otimes \sum_n |n\rangle\langle n|) \cdot \\ &\quad V_l \cdot (\mathbf{1}_S \otimes |m\rangle) \cdot \rho_S(t) \\ &\quad - \frac{1}{2} \sum_{l,m} \lambda_m \rho_S(t) \cdot (\mathbf{1}_S \otimes \langle m|) V_l^\dagger \cdot (\mathbf{1}_S \otimes \sum_n |n\rangle\langle n|) \\ &\quad \cdot V_l \cdot (\mathbf{1}_S \otimes |m\rangle) \end{aligned} \quad (121)$$

By defining the operators

$$c_{l,n,m} = \sqrt{\frac{\lambda_m}{dt}} (\mathbf{1}_S \otimes \langle n|) \cdot V_l \cdot (\mathbf{1}_S \otimes |m\rangle), \quad (122)$$

we end up with the Lindblad operator

$$\frac{\partial \rho_S}{\partial t} = \sum_{l,n,m} c_{l,n,m} \rho_S c_{l,n,m}^\dagger - \frac{1}{2} \left(\rho_S c_{l,n,m}^\dagger c_{l,n,m} + c_{l,n,m}^\dagger c_{l,n,m} \rho_S \right), \quad (123)$$

up to a reindexation. Since the Lindblad operator derives from a completely positive and trace preserving map $\mathcal{V}$ (115), it effectively describes a physical evolution as the density operator will remain positive and of unit trace under this evolution (123). A more general and detailed derivation of the Lindblad operator is given in [45].

Appendix B Clebsch-Gordan Coefficients

In this appendix we detail the tables of the Clebsch-Gordan coefficients

$$\langle j_1, j_2; m_1, m_2 | J, M \rangle \quad (124)$$

that we use in this work. Based on [46] we recall the three relations

$$\langle j_1, j_2; m_1, m_2 | J, M \rangle = 0 \text{ if } M \neq m_1 + m_2, \quad (125)$$

$$\langle j_1, j_2; m_1, m_2 | J, M \rangle = (-1)^{j_1+j_2-J} \langle j_2, j_1; m_2, m_1 | J, M \rangle \quad (126)$$

$$\langle j_1, j_2; m_1, m_2 | J, M \rangle = (-1)^{j_1+j_2+J} \langle j_2, j_1; -m_2, -m_1 | J, -M \rangle \quad (127)$$

Such that we focus on the cases where $M \geq 0$ and $j_1 \geq j_2$. In addition, all the transitions that we consider satisfy the condition

$$J = j_1 + j_2 \quad (128)$$

The simplest case involves the 4^1S_0 manifold, such that $j_2 = 0$ and

$$\langle j_1, 0; m_1, 0 | J, M \rangle = \delta_{J=j_1} \delta_{M=m_1} \quad (129)$$

For the case involving the 4^1P_1 and 4^1D_2 states, we need the coefficients in Table 8

M	2	1		0		
(m_1, m_2)	(1, 1)	(1, 0)	(0, 1)	(1, -1)	(0, 0)	(-1, 1)
$\langle 1, 1; m_1, m_2 2, M \rangle$	1	$\sqrt{\frac{1}{2}}$	$\sqrt{\frac{1}{2}}$	$\sqrt{\frac{1}{6}}$	$\sqrt{\frac{2}{3}}$	$\sqrt{\frac{1}{6}}$

Table 8: Values of the Clebsch-Gordan coefficients for $(j_1, j_2, J) = (1, 1, 2)$, taken from [46].

Appendix C Polarization Spectroscopy

C.1 Output signal

We assume that the probe beam after the first polarization beam splitter, see Figure 6, is polarized along the x axis so that its complex field can be decomposed into two circular polarization components

$$\vec{E}^{(+)}(z, t) = \frac{E_0}{2} e^{-i(\omega t - kz)} \vec{e}_x = \frac{E_0}{2\sqrt{2}} e^{-i(\omega t - kz)} (-\vec{\epsilon}_+ + \vec{\epsilon}_-). \quad (130)$$

$\vec{\epsilon}_\pm = \mp \frac{\vec{e}_x \pm i\vec{e}_y}{\sqrt{2}}$ correspond to the left and right circular polarization vectors.

Now suppose that as the right (left) circular polarization passes through the lamp of length L , it encounters a complex refractive index n_+ (n_-) due to the interaction with the atoms in the optogalvanic lamp and another complex refractive index w_+ (w_-) due to the windows of the lamp. Then, we can define the quantities

$$\begin{aligned} n_{tot} &= \frac{n_+ + n_-}{2} & \text{and} & \quad \Delta n = n_+ - n_- \\ w_{tot} &= \frac{w_+ + w_-}{2} & \text{and} & \quad \Delta w = w_+ - w_- \end{aligned} \quad (131)$$

The resulting field at the output of the lamp is

$$\begin{aligned} \vec{E}_{out}^{(+)} &= \frac{E_0}{2\sqrt{2}} \left(-e^{-i(\omega t - (n_+ + w_+) \frac{\omega}{c} L)} \vec{\epsilon}_+ + e^{-i(\omega t - (n_- + w_-) \frac{\omega}{c} L)} \vec{\epsilon}_- \right) \\ &= \frac{E_0}{2\sqrt{2}} e^{-i(\omega t - (n_{tot} + w_{tot}) \frac{\omega}{c} L)} \left(-e^{i(\Delta n + \Delta w) \frac{\omega}{2c} L} \vec{\epsilon}_+ + e^{-i(\Delta n + \Delta w) \frac{\omega}{2c} L} \vec{\epsilon}_- \right). \end{aligned} \quad (132)$$

As shown in Figure 6, after the optogalvanic lamp, the probe beam passes through a half-wave plate. Assuming that the fast axis of the wave plate is rotated by an angle θ compared to the x axis, the effect of this half-wave plate on the polarization axis of the probe beam using Jones calculus [47], is described by the matrix

$$M_{HWP}(\theta) = -i \begin{pmatrix} \cos 2\theta & \sin 2\theta \\ \sin 2\theta & -\cos 2\theta \end{pmatrix}, \quad (133)$$

in the $\{x, y\}$ basis. Then, with the second polarization beam splitter, see Figure 6, the field is projected onto each linear component x and y . Taking into account the effect of the half-wave plate, the two components going to the inputs of the balanced photo-detector are

$$\begin{aligned} E_x^{(+)} &= -i \frac{E_0}{4} e^{-i(\omega t - (n_{tot} + w_{tot}) \frac{\omega}{c} L)} \left(e^{i(\Delta n + \Delta w) \frac{\omega}{2c} L} e^{i2\theta} + e^{-i(\Delta n + \Delta w) \frac{\omega}{2c} L} e^{-i2\theta} \right) \\ E_y^{(+)} &= -\frac{E_0}{4} e^{-i(\omega t - (n_{tot} + w_{tot}) \frac{\omega}{c} L)} \left(e^{i(\Delta n + \Delta w) \frac{\omega}{2c} L} e^{i2\theta} - e^{-i(\Delta n + \Delta w) \frac{\omega}{2c} L} e^{-i2\theta} \right). \end{aligned} \quad (134)$$

Eventually, since the intensity is the square modulus of the incoming field and the balanced photo-detector returns the difference of the two intensities, we obtain the signal

$$\Delta I = \frac{E_0^2}{2} e^{-2\mathcal{I}m(n_{tot}+w_{tot})\frac{\omega}{c}L} \cos\left(\mathcal{R}e(\Delta n + \Delta w)\frac{\omega}{c}L - 2\theta\right). \quad (135)$$

Choosing the angle between the x axis, which is the initial polarization axis of the probe beam, and the fast axis of the half-wave plate to be

$$\theta = \frac{\pi}{4} + \mathcal{R}e(\Delta w)\frac{\omega}{2c}L \quad (136)$$

and assuming that $\mathcal{R}e(\Delta n)\frac{\omega}{c}L$ is small, the final signal is

$$\Delta I = \frac{E_0^2}{2} e^{-2\mathcal{I}m(n_{tot}+w_{tot})\frac{\omega}{c}L} \mathcal{R}e(\Delta n)\frac{\omega}{c}L. \quad (137)$$

We can get the value (136), for the angle θ between the initial polarization axis of the probe beam and the fast axis of the last half-wave plate, by adjusting the half wave plate to have an output signal equal to zero when the lamp is off. Indeed, in this case, the real part of the difference of the refractive indices is zero as well as the output signal (135) when the half wave plate is rotated by the angle (136).

C.2 Difference of refractive indices

In this subsection, we calculate the real part of the difference of refractive indices in the polarization spectroscopy setup for the usual case where the probe beam intensity is weak relative to the saturation intensity (34). The reference we use for the calculation is [48] leading to the analytical expression in [21]. The ground state 4^1S_0 and the excited state 4^1P_1 , see Figure 7, would be referred as state $|1\rangle$ and state $|2\rangle$. When the distinction between the two sub-levels of interest in the 4^1P_1 Zeeman manifold is required, they will be differentiated by the superscripts $+$ and $-$.

The linewidth of the transition when considering only the saturating pump beam is γ_s . In the case of the weak probe beam, we can neglect the Rabi frequency with respect to the linewidth $|\Omega_{probe}| \ll \gamma_{12}$. Thus, the linear dielectric susceptibility (40) becomes

$$\chi = \frac{\Delta N}{V} \frac{|d_{12}|^2}{\hbar\epsilon_0} \frac{1}{2} \frac{\delta + i\gamma_{12}}{\delta^2 + \gamma_{12}^2} \quad \text{with} \quad \Delta N = N(\rho_{11}^0 - \rho_{22}^0). \quad (138)$$

$N\rho_{ii}^0$ is the population in level i neglecting the weak probe beam (36). Indeed, given the assumption that the probe beam is weak, it will barely affect

the atomic medium traversed by the pump beam. Therefore, as the pump beam saturates the $m_0 = 0 \leftrightarrow m_1 = +1$ transition, this results in different population differences denoted by ΔN^+ and ΔN^- . The population differences, expressed with the saturation parameter s (33) of the pump beam, are

$$\begin{aligned}\Delta N^+ &= N(\rho_{11} - \rho_{22}^+) = N(1 - \frac{s}{1+s}) \\ \Delta N^- &= N(\rho_{11} - \rho_{22}^-) = N(1 - \frac{1}{2} \frac{s}{1+s})\end{aligned}\quad (139)$$

Thus, using the expression for the saturation parameter (33), the relations (138) and (139), and taking into account the Doppler shifts for the two counter-propagating beams, the difference of refractive indices in the polarization spectroscopy setup is

$$\begin{aligned}\Delta n &= \frac{|d_{12}|^2}{2\hbar\epsilon_0} \int \frac{dv}{\sqrt{2\pi v_{th}^2}} e^{-\frac{v^2}{2v_{th}^2}} \frac{1}{2} \frac{\delta - kv + i\gamma_{12}}{(\delta - kv)^2 + \gamma_{12}^2} \cdot \frac{\Delta N^+ - \Delta N^-}{V} \\ &= -\frac{N}{V} \frac{|d_{12}|^2}{8\hbar\epsilon_0} \int \frac{dv}{\sqrt{2\pi v_{th}^2}} e^{-\frac{v^2}{2v_{th}^2}} \frac{1}{(\delta - kv) - i\gamma_{12}} \cdot \frac{\frac{\Omega^2}{2} \frac{2\gamma_{12}}{\Gamma_{12}}}{(\delta + kv)^2 + \gamma_s^2}\end{aligned}\quad (140)$$

We can neglect the sign in front of the expression (140) because it could be reversed by rotating the half-wave plate in front of the second polarization beam splitter, see Figure 6, by an angle $\Delta\theta = \frac{\pi}{2}$ in (135).

As the gaussian varies over a typical scale much larger than the other two functions ($kv_{th} \gg \gamma_{12}, \gamma_s$), we can consider it as a constant and integrate the remaining function with the residue theorem. As of the three poles of the function, $\delta - i\gamma_{12}$ and $-\delta \pm i\gamma_s$, only one has a positive imaginary part. As a consequence, we consider a closed integral path around it. The remaining function to be integrated, thus not taking into account the gaussian, converges to zero quickly enough for the integral (140) to be equal to the residue at $-\delta + i\gamma_s$.

$$\begin{aligned}\Delta n &= -\frac{N}{V} \frac{|d_{12}|^2}{8\hbar\epsilon_0} e^{-\frac{\delta^2}{2(kv_{th})^2}} \frac{1}{\sqrt{2\pi}kv_{th}} \frac{1}{2\delta - i(\gamma_{12} + \gamma_s)} \cdot \frac{2\pi i \frac{\Omega^2}{2} \frac{2\gamma_{12}}{\Gamma_{12}}}{2i\gamma_s} \\ &= -\frac{N}{V} \frac{|d_{12}|^2}{8\hbar\epsilon_0} \frac{\Omega^2 \gamma_{12}}{2\Gamma_{12}} \frac{\pi}{\sqrt{2\pi}kv_{th}\gamma_s} e^{-\frac{\delta^2}{2(kv_{th})^2}} \frac{\delta + i\frac{1}{2}(\gamma_{12} + \gamma_s)}{\delta^2 + \frac{1}{4}(\gamma_{12} + \gamma_s)^2}\end{aligned}\quad (141)$$

Finally, the output signal of the balanced photo-detector is proportional to the real part of the difference of refractive indices, which is

$$\mathcal{Re}(\Delta n) = -\frac{N}{V} \frac{|d_{12}|^2}{8\hbar\epsilon_0} \frac{\Omega^2 \gamma_{12}}{2\Gamma_{12}} \frac{\pi}{\sqrt{2\pi}kv_{th}\gamma_s} e^{-\frac{\delta^2}{2(kv_{th})^2}} \frac{\delta}{\delta^2 + \frac{1}{4}(\gamma_{12} + \gamma_s)^2}.\quad (142)$$

This expression (142), which is plotted in Figure 33, has a dispersion-like shape, which is then also the case for the output signal (137) and centered

on the atomic resonance or zero detuning. Due to this property, we can use the polarization spectroscopy technique to lock the laser frequency to the resonance. Furthermore, since the signal (142) in Figure 33 is plotted for different Rabi frequencies of the pump beam, we see the power broadening effect. Indeed, the higher the Rabi frequency, the further apart the extrema of the curves are. Nevertheless, as the Rabi frequency also changes the signal amplitude, a lower Rabi frequency is not tantamount to a steeper signal as can be seen in Figure 33.

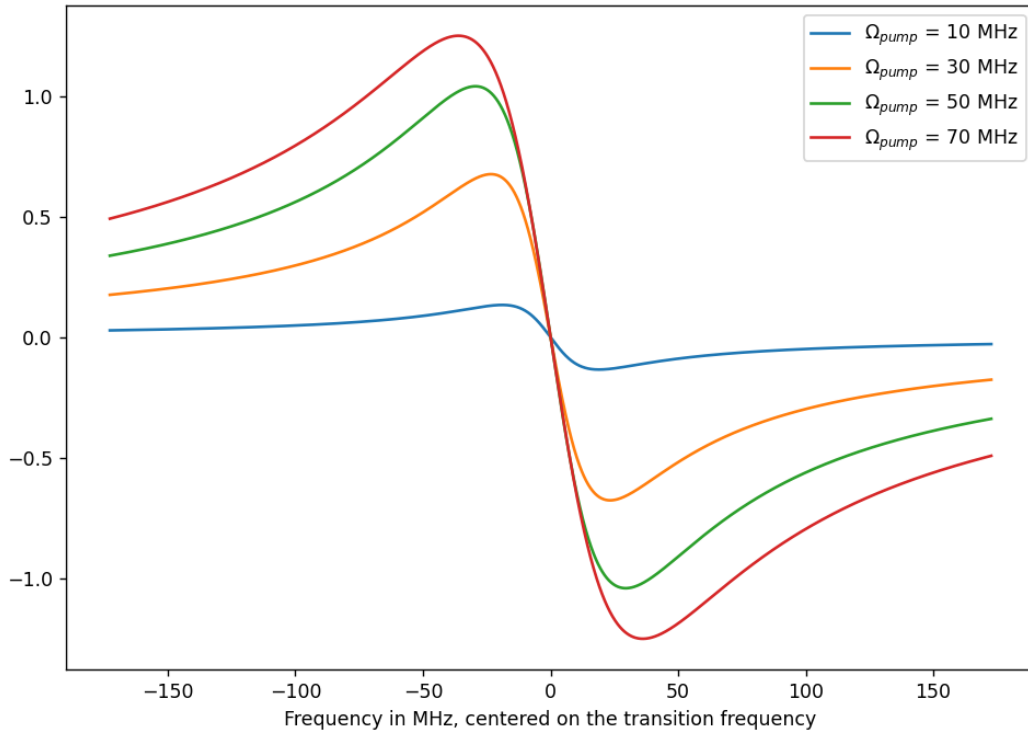


Figure 33: Real part of the difference of refractive indices $\mathcal{Re}(\Delta n)$ in the polarization spectroscopy experiment. For different values of the pump Rabi frequency Ω_{pump} . Decay rate $\Gamma_{12} = 34.5$ MHz, dephasing rates $\gamma_{coll} + \gamma_{las} = 1$ MHz, wavelength $\lambda = 423$ nm and temperature $T = 600$ K.

Appendix D Technicalities

In this appendix, we detail the different devices such as the lock-in amplifier and the acousto-optic modulator that we use in our experiments to be able to filter out noise.

D.1 Lock in amplifier

In all of our experiments, polarization spectroscopy, observation of the absorption drop in a three-level ladder system, and two-color polarization spectroscopy, we use lock-in amplifiers to filter out noise added to the signal. Specifically, we typically use a square wave signal from a waveform generator and a switch to chop the control beam at a typical frequency of 100 kHz and create an internal reference signal for the lock-in amplifier at the same frequency, as shown on Figure 34. Since the quantities governing the evolution of the optical experiments, the Rabi frequencies and the spontaneous decay rates, are about 1 – 10 MHz, we can still make the approximation that despite the chopped excitation at 100 kHz, the atoms still reach their steady state. In this approximation, the signal going to the photo-detectors is modulated and alternates between the one we get in the steady state regime when the control beam is blocked and the one when it is not. Note that the bandwidths of the photo-detectors that we use are also higher than 100 kHz, in the case of the *Thorlabs* model *PDA36A-EC* it is 10 MHz and for the *Nirvana detector* model *2007* it is 125 kHz. The output signal of the photo-detector is then sent to the lock-in amplifier, which will act as a bandpass filter centered on the reference frequency 100 kHz allowing to suppress noise at different frequencies and to eventually amplify the filtered signal.

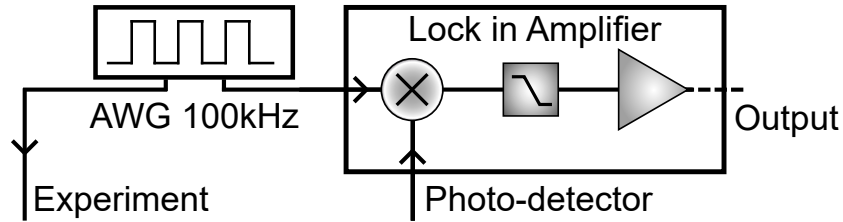


Figure 34: Illustration of a lock in amplifier fed by an arbitrary waveform generator (AWG).

To describe more mathematically the operating principle of a lock-in amplifier, we rely on the detailed note [49]. The lock-in amplifier is fed with an external reference signal, coming from a waveform generator in our case,

such as a square wave signal at angular frequency ω_{ref} . From this signal, the lock-in amplifier generates an internal sinusoidal reference signal

$$U_{reference}(t) = V_{ref} \sin(\omega_{ref}t + \theta_{ref}). \quad (143)$$

By considering the component of the signal at the angular frequency ω_{sig}

$$U_{signal}(t) = V_{sig} \sin(\omega_{sig}t + \theta_{sig}), \quad (144)$$

the lock-in amplifier multiplies it with its internal reference signal (143) as shown in Figure 34. The resulting signal is

$$U_{output}(t) = \frac{1}{2} V_{ref} V_{sig} \{ \cos([\omega_{ref} - \omega_{sig}]t + [\theta_{ref} - \theta_{sig}]) - \cos([\omega_{ref} + \omega_{sig}]t + [\theta_{ref} + \theta_{sig}]) \}. \quad (145)$$

Since the signal of interest is chopped at the same frequency as the reference signal of the lock-in amplifier, we wish to retain only the DC component of the output signal (145). As a consequence, a low-pass filter is applied to keep the low-frequency components of the photo-detector signal (145) which are finally amplified, see Figure 34. Therefore, the lock-in amplifier acts as a band-pass filter around its reference frequency and in our case, it allows to attenuate noise at frequencies different from 100 kHz.

There are three main parameters of the lock-in amplifier that we can adjust:

- The phase of the internal reference signal θ_{ref} . Indeed, we can see that the resulting DC component of the output signal (145) is proportional to the cosine of the difference between this phase and that of the signal θ_{sig} .
- The time constant which defines the cutoff frequency of the low-pass filter.
- The sensitivity which defines the gain of the lock-in amplifier.

D.2 Acousto-optic modulator

Acousto-optic modulators are electronic devices that contain a medium, such as quartz for instance, whose refractive index n changes when it is subjected to mechanical stress by a piezoelectric transducer [50]. The piezo is driven by a periodic radio frequency (RF) signal of frequency f_{RF} . Due to the electro-mechanical properties of the piezo, a standing acoustic wave is

created in the medium and a modulation of the refractive index occurs. This modulation has the same periodicity as the applied RF signal and we define its wavelength as Λ . When passing through the acousto-optics modulator, the light is subject to Bragg diffraction. The light of wavelength λ and frequency ν arriving with the Bragg angle

$$\theta_B = \frac{\lambda}{2n\Lambda}, \quad (146)$$

in the medium will come out in possibly different orders, the m^{th} order being separated of the 0^{th} order by $2m$ times the Bragg angle (146). In this case, it means that the light has interacted with m phonons inside the acousto-optics modulator and therefore its frequency will be shifted by the quantity [51]

$$\Delta\nu = m \times f_{RF}. \quad (147)$$

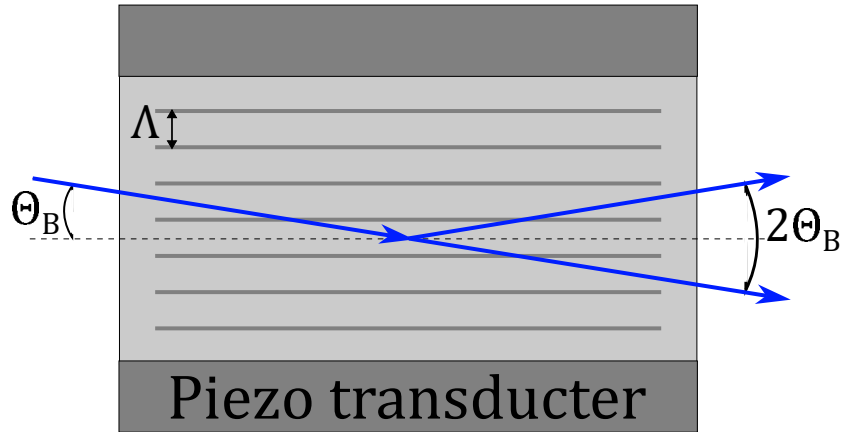


Figure 35: Illustration of a beam light interacting with an acousto-optics modulator resulting in both a transmitted and a first order diffracted beam. Θ_B is the Bragg angle, Λ is the wavelength of the sound wave in the medium.

A calculation [51] gives the intensity of the first order compared to the beam intensity

$$I_1 = I_0 \sin^2 \left(\alpha \sqrt{P} \right), \quad (148)$$

where I_0 is the input power of the light beam and P is the power of the applied RF signal applied. Eventually, in our experiments we use acousto-optics modulators to replace mechanical choppers. To do so, we modulate the input RF signal with a switch and a square wave signal at $f_{mod} = 100$ kHz generated by an arbitrary waveform generator. When the square signal is zero, the light is not diffracted and is blocked by a beam wall after the

acousto-optic modulator. When the square signal is on, the light is diffracted and its trajectory is modified by an angle $2\theta_B$. It then avoids the beam block and continues to the experiment. Note that using an acousto-optics modulator instead of a mechanical device to chop the signal allows to use higher frequencies, for instance mechanical choppers can go up to 10 kHz, though it can also induce a detuning in spectroscopy techniques.

Appendix E Three-Level Ladder System

In this appendix, we focus on solving the optical Bloch equations (22) for the hamiltonian (74) describing a three-level ladder system and the Lindblad operator (75) describing the incoherent processes.

Strictly speaking, there are nine equations contained in the matrix equation (22), however they can be summarized into five ones by taking into account the hermitian character of the density matrix operator, i.e. $\rho_{i,j} = \rho_{j,i}^*$, and its unit trace, i.e. $\rho_{11} + \rho_{22} + \rho_{33} = 1$ implying $\dot{\rho}_{11} + \dot{\rho}_{22} + \dot{\rho}_{33} = 0$. We recall that we consider real Rabi frequencies and that we use the quantities γ_{ij} defined in the expression (76).

$$\begin{cases} \dot{\rho}_{11} &= \Gamma_{12}\rho_{22} + \Gamma_{13}\rho_{33} + i\frac{\Omega_p}{2}(\rho_{12} - \rho_{21}) \\ \dot{\rho}_{33} &= -(\Gamma_{13} + \Gamma_{23})\rho_{33} + i\frac{\Omega_c}{2}(\rho_{32} - \rho_{23}) \\ \dot{\rho}_{12} &= -(\gamma_{12} - i\Delta_p)\rho_{12} + i\frac{\Omega_c}{2}\rho_{13} + i\frac{\Omega_p}{2}(\rho_{11} - \rho_{22}) \\ \dot{\rho}_{13} &= -(\gamma_{13} - i(\Delta_p + \Delta_c))\rho_{13} + i\frac{\Omega_c}{2}\rho_{12} - i\frac{\Omega_p}{2}\rho_{23} \\ \dot{\rho}_{23} &= -(\gamma_{23} - i\Delta_c)\rho_{23} - i\frac{\Omega_p}{2}\rho_{13} + i\frac{\Omega_c}{2}(\rho_{22} - \rho_{33}) \end{cases} \quad (149)$$

In this appendix, we consider only the steady state regime and we therefore set the time derivatives to zero.

E.1 Limit $\Omega_p \ll \Omega_c, \Gamma_{12}$

In this subsection, we solve the optical Bloch equations (149) describing the three-level ladder system in the limit where the intensity of the control beam is much higher than the one of the probe beam

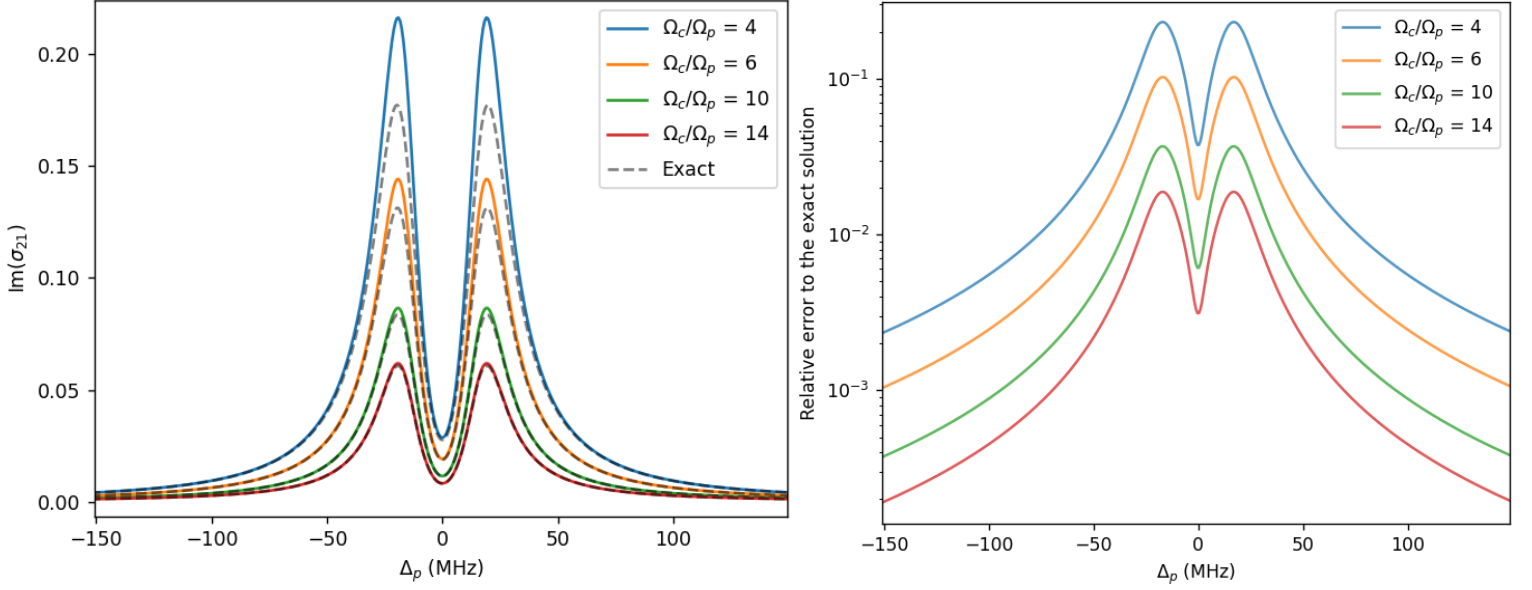
$$\Omega_p \ll \Omega_c, \Gamma_{12} \quad (150)$$

Following the article [25], simple theoretical expressions for the steady state solutions of ρ_{12} and ρ_{13} can be found under this assumptions. Indeed, this means that the probe beam coupling atoms initially in the ground state $|1\rangle$ to other states is relatively weak compare to the other processes. As a consequence, the initial coefficients of the density matrix $\rho_{ij}^{(0)} = \delta_{i=j=1}$ change only slightly to $\rho_{ij}^{(0)} + \rho_{ij}^{(1)}$. Then, equations (149.3) and (149.4) form a close system for $\rho_{12}^{(1)}$ and $\rho_{13}^{(1)}$ because we neglect the lower order terms like $\Omega_p \rho_{ij}^{(1)}$

$$\begin{aligned} 0 &= -(\gamma_{12} - i\Delta_p)\rho_{12}^{(1)} + i\frac{\Omega_c}{2}\rho_{13}^{(1)} + i\frac{\Omega_p}{2} \\ 0 &= -(\gamma_{13} - i(\Delta_p + \Delta_c))\rho_{13}^{(1)} + i\frac{\Omega_c}{2}\rho_{12}^{(1)} \end{aligned} \quad (151)$$

To simplify and since $\rho_{12}^{(0)} = \rho_{13}^{(0)} = 0$ we do not write the exponent ⁽¹⁾ in the rest of this subsection. The solutions are

$$\begin{aligned}\rho_{12} &= i \frac{\Omega_p}{2} \frac{\gamma_{13} - i(\Delta_p + \Delta_c)}{(\gamma_{13} - i(\Delta_p + \Delta_c))(\gamma_{12} - i\Delta_p) + \frac{\Omega_c^2}{4}} \\ \rho_{13} &= -\frac{\Omega_p \Omega_c}{4} \frac{1}{(\gamma_{13} - i(\Delta_p + \Delta_c))(\gamma_{12} - i\Delta_p) + \frac{\Omega_c^2}{4}}\end{aligned}\quad (152)$$



(a) Theoretical expression [25] of $\text{Im}(\rho_{21})$

(b) Relative error of the analytical solution

Figure 36: Comparison of $\text{Im}(\rho_{21})$ from the expression (152) and the exact solution, for different Rabi frequencies of the probe beam. Control Rabi frequency $\Omega_c = 37.4$ MHz, transition linewidth $(\Gamma_{12}, \Gamma_{23}, \Gamma_{13}) = (34.5, 2.5, 0.35)$ MHz, dephasing rates $\gamma_p = \gamma_c = 1$ MHz and a control beam on resonance $\Delta_c = 0$

In Figure 36 (a) we plot the imaginary part of the coherent term ρ_{21} for the simplified expression (152) in color and the exact solution, in dashed black, using a matrix solver. We fix the control Rabi frequency and use different probe Rabi frequencies. In Figure 36 (b) we calculate the relative error between the simplified expression (152) and the exact expression. Not surprisingly, the higher the ratio $\frac{\Omega_c}{\Omega_p}$, the smaller the relative error between the exact expression and simplified expression. When the ratio $\frac{\Omega_c}{\Omega_p}$ is close to 1 then the approximation (150) collapses and we obtain important relative error. However, in the experiments we conduct, we sometimes work with ratios close to 1. This is why we develop a more accurate expression in

the next subsection. This expression would be independent of the ratio of the Rabi frequencies and would assume only a small decay rate from state $|3\rangle$ to state $|1\rangle$ compared to the others typical frequencies of the problem. This approximation is reasonable in our case because we consider the neutral calcium atoms whose state $|1\rangle$ is 4^1S_0 and whose state $|3\rangle$ is 4^1D_2 , so they are only directly coupled by quadrupole transition.

E.2 Approximated analytical expression

Returning to the optical Bloch equations (149), in this subsection we derive an analytical expression for $\mathcal{I}m(\rho_{21})$ in the steady state regime. To do this, we first neglect the two terms with Γ_{13} in the equations (149), as this decay rate is much smaller than any other decay rates or Rabi frequencies for the levels of neutral calcium atoms we consider. We use the notations defined in (76) and introduce the two-photon detuning $\Delta = \Delta_p + \Delta_c$. The equations (149) can then be rewritten with the real and imaginary parts of the coefficients of the density matrix

$$\left\{ \begin{array}{lcl} \Omega_p \mathcal{I}m(\rho_{12}) & = & \Gamma_{12} \rho_{22} \\ \Omega_c \mathcal{I}m(\rho_{23}) & = & \Gamma_{23} \rho_{33} \\ \gamma_{23} \mathcal{R}e(\rho_{23}) & = & \frac{\Omega_p}{2} \mathcal{I}m(\rho_{13}) - \Delta_c \mathcal{I}m(\rho_{23}) \\ \gamma_{13} \mathcal{I}m(\rho_{13}) - \Delta \mathcal{R}e(\rho_{13}) & = & \frac{\Omega_c}{2} \mathcal{R}e(\rho_{12}) - \frac{\Omega_p}{2} \mathcal{R}e(\rho_{23}) \\ \frac{\Omega_p}{2} \mathcal{R}e(\rho_{13}) - \frac{\Omega_c}{2} (\rho_{22} - \rho_{33}) & = & \Delta_c \mathcal{R}e(\rho_{23}) - \gamma_{23} \mathcal{I}m(\rho_{23}) \\ \gamma_{12} \mathcal{R}e(\rho_{12}) & = & -\frac{\Omega_c}{2} \mathcal{I}m(\rho_{13}) - \Delta_p \mathcal{I}m(\rho_{12}) \\ -\frac{\Omega_p}{2} (\rho_{11} - \rho_{22}) - \Delta_p \mathcal{R}e(\rho_{12}) & = & \frac{\Omega_c}{2} \mathcal{R}e(\rho_{13}) - \gamma_{12} \mathcal{I}m(\rho_{12}) \\ \gamma_{13} \mathcal{R}e(\rho_{13}) + \Delta \mathcal{I}m(\rho_{13}) & = & -\frac{\Omega_c}{2} \mathcal{I}m(\rho_{12}) + \frac{\Omega_p}{2} \mathcal{I}m(\rho_{23}) \end{array} \right. \quad (153)$$

The unitary trace relation is not written for simplicity. In the following, we will refer to these equations (153) as (153.i) for $i \in \{1 \text{ to } 8\}$. For instance, with equation (153.1), we see that it is sufficient to have an expression for ρ_{22} to determine $\mathcal{I}m(\rho_{21})$. Replacing $\mathcal{R}e(\rho_{23})$ with equation (153.3), the equations (153.4) and (153.5) become

$$\left\{ \begin{array}{l} \left(\gamma_{23} \gamma_{13} + \left(\frac{\Omega_p}{2} \right)^2 \right) \mathcal{I}m(\rho_{13}) - \gamma_{23} \Delta \mathcal{R}e(\rho_{13}) = \\ \quad \frac{\Omega_p}{2} \Delta_c \mathcal{I}m(\rho_{23}) + \gamma_{23} \frac{\Omega_c}{2} \mathcal{R}e(\rho_{12}) \\ \frac{\Omega_p}{2} \Delta_c \mathcal{I}m(\rho_{13}) - (\gamma_{23}^2 + \Delta_c^2) \mathcal{I}m(\rho_{23}) = \\ \quad \gamma_{23} \frac{\Omega_p}{2} \mathcal{R}e(\rho_{13}) - \gamma_{23} \frac{\Omega_c}{2} (\rho_{22} - \rho_{33}) \end{array} \right. \quad (154)$$

Then, replacing $\mathcal{R}e(\rho_{12})$ by its expression equation (153.6) as well as $\mathcal{I}m(\rho_{12})$ and $\mathcal{I}m(\rho_{23})$ thanks to the equations (153.1) and (153.2), we obtain the

system of four equations

$$\left\{ \begin{array}{l} \gamma_{23} \frac{\Omega_c}{2} \rho_{22} - \left(\gamma_{23} \frac{\Omega_c}{2} + \frac{\Gamma_{23}}{\Omega_c} (\gamma_{23}^2 + \Delta_c^2) \right) \rho_{33} = \\ \quad \gamma_{23} \frac{\Omega_p}{2} \mathcal{R}e(\rho_{13}) - \frac{\Omega_p}{2} \Delta_c \mathcal{I}m(\rho_{13}) \\ \left(\gamma_{23} \gamma_{13} + \left(\frac{\Omega_p}{2} \right)^2 + \frac{\gamma_{23}}{\gamma_{12}} \left(\frac{\Omega_c}{2} \right)^2 \right) \mathcal{I}m(\rho_{13}) - \gamma_{23} \Delta \mathcal{R}e(\rho_{13}) = \\ \quad \frac{\Gamma_{23}}{2} \frac{\Omega_p}{\Omega_c} \Delta_c \rho_{33} - \frac{\Gamma_{12}}{2\gamma_{12}} \frac{\Omega_c}{\Omega_p} \gamma_{23} \Delta_p \rho_{22} \\ - \frac{\Omega_p}{2} (1 - 2\rho_{22} - \rho_{33}) + \frac{\Gamma_{12}}{\Omega_p} \left(\gamma_{12} + \frac{\Delta_p^2}{\gamma_{12}} \right) \rho_{22} = \\ \quad \frac{\Omega_c}{2} \mathcal{R}e(\rho_{13}) - \frac{\Omega_c}{2} \frac{\Delta_p}{\gamma_{12}} \mathcal{I}m(\rho_{13}) \\ \gamma_{13} \mathcal{R}e(\rho_{13}) + \Delta \mathcal{I}m(\rho_{13}) = - \frac{\Gamma_{12}}{2} \frac{\Omega_c}{\Omega_p} \rho_{22} + \frac{\Gamma_{23}}{2} \frac{\Omega_p}{\Omega_c} \rho_{33} \end{array} \right. \quad (155)$$

The last equation (155.4) allows to replace $\mathcal{R}e(\rho_{13})$ in the previous expressions. By doing this and using the equation (155.2) we obtain an expression for $\mathcal{I}m(\rho_{13})$

$$\mathcal{I}m(\rho_{13}) = \frac{\frac{\Gamma_{23}}{2} \frac{\Omega_p}{\Omega_c} \left(\frac{\Delta_c}{\gamma_{23}} + \frac{\Delta}{\gamma_{13}} \right) \rho_{33} - \frac{\Gamma_{12}}{2} \frac{\Omega_c}{\Omega_p} \left(\frac{\Delta_p}{\gamma_{12}} + \frac{\Delta}{\gamma_{13}} \right) \rho_{22}}{\gamma_{13} + \frac{1}{\gamma_{23}} \left(\frac{\Omega_p}{2} \right)^2 + \frac{1}{\gamma_{12}} \left(\frac{\Omega_c}{2} \right)^2 + \frac{\Delta^2}{\gamma_{13}}}. \quad (156)$$

Finally, by defining the positive quantities

$$\Gamma = \gamma_{13} + \frac{\Omega_p^2}{4\gamma_{23}} + \frac{\Omega_c^2}{4\gamma_{12}} + \frac{\Delta^2}{\gamma_{13}}, \quad \alpha = \left(\frac{\Delta_c}{\gamma_{23}} + \frac{\Delta}{\gamma_{13}} \right), \quad \beta = \left(\frac{\Delta_p}{\gamma_{12}} + \frac{\Delta}{\gamma_{13}} \right), \quad (157)$$

we end up with two equations linking ρ_{22} and ρ_{33}

$$\left\{ \begin{array}{l} \left(1 + \frac{\Gamma_{23}}{2\gamma_{23}} \left(\frac{2}{\Omega_c} \right)^2 (\gamma_{23}^2 + \Delta_c^2) + \left(\frac{\Omega_p}{\Omega_c} \right)^2 \left(\frac{\Gamma_{23}}{2\gamma_{13}} - \frac{\Gamma_{23}}{2\Gamma} \alpha^2 \right) \right) \rho_{33} = \\ \quad \left(1 + \frac{\Gamma_{12}}{2\gamma_{13}} - \frac{\Gamma_{12}}{2\Gamma} \alpha \beta \right) \rho_{22} \\ \left(2 + \left(\frac{2}{\Omega_p} \right)^2 \left(\frac{\Gamma_{12}}{2\gamma_{12}} (\gamma_{12}^2 + \Delta_p^2) + \frac{\Gamma_{12}}{2\gamma_{13}} \left(\frac{\Omega_c}{2} \right)^2 - \frac{\Gamma_{12}}{2\Gamma} \beta^2 \left(\frac{\Omega_c}{2} \right)^2 \right) \right) \rho_{22} = \\ \quad 1 - \left(1 - \frac{\Gamma_{23}}{2\gamma_{13}} + \frac{\Gamma_{23}}{2\Gamma} \alpha \beta \right) \rho_{33} \end{array} \right. \quad (158)$$

Using the saturation parameters (33) for both beams

$$s_p = \frac{\Omega_p^2}{2} \frac{2\gamma_{12}}{\Gamma_{12}} \frac{1}{\Delta_p^2 + \gamma_{12}^2} \quad \text{and} \quad s_c = \frac{\Omega_c^2}{2} \frac{2\gamma_{23}}{\Gamma_{23}} \frac{1}{\Delta_c^2 + \gamma_{23}^2}, \quad (159)$$

we obtain the condensed expressions

$$\left\{ \begin{array}{l} \left(1 + \frac{2}{s_c} + \frac{\Omega_p^2}{\Omega_c^2} \left(\frac{\Gamma_{23}}{2\gamma_{13}} - \frac{\Gamma_{23}}{2\Gamma} \alpha^2 \right) \right) \rho_{33} = \left(1 + \frac{\Gamma_{12}}{2\gamma_{13}} - \frac{\Gamma_{12}}{2\Gamma} \alpha \beta \right) \rho_{22} \\ 1 - \left(1 - \frac{\Gamma_{23}}{2\gamma_{13}} + \frac{\Gamma_{23}}{2\Gamma} \alpha \beta \right) \rho_{33} = \left(2 + \frac{2}{s_p} + \frac{\Omega_c^2}{\Omega_p^2} \left(\frac{\Gamma_{12}}{2\gamma_{13}} - \frac{\Gamma_{12}}{2\Gamma} \beta^2 \right) \right) \rho_{22} \end{array} \right. \quad (160)$$

A sanity check is to consider that there is no control field, so as $\rho_{33} = 0$ and $\Omega_c = 0$, the equation (160.2) gives us the same population of the excited state (36) as in the case of a two-level system.

Eventually, we get the final expression for ρ_{22}

$$\left\{ \begin{array}{l} \frac{\rho_{33}}{\rho_{22}} = \frac{1 + \frac{\Gamma_{12}}{2\gamma_{13}} - \frac{\Gamma_{12}}{2\Gamma} \alpha\beta}{1 + \frac{2}{s_c} + \frac{\Omega_p^2}{\Omega_c^2} \left(\frac{\Gamma_{23}}{2\gamma_{13}} - \frac{\Gamma_{23}}{2\Gamma} \alpha^2 \right)} \\ \frac{1}{\rho_{22}} = 2 + \frac{2}{s_p} + \frac{\Omega_c^2}{\Omega_p^2} \left(\frac{\Gamma_{12}}{2\gamma_{13}} - \frac{\Gamma_{12}}{2\Gamma} \beta^2 \right) + \left(1 - \frac{\Gamma_{23}}{2\gamma_{13}} + \frac{\Gamma_{23}}{2\Gamma} \alpha\beta \right) \frac{\rho_{33}}{\rho_{22}} \end{array} \right. \quad (161)$$

We can then easily get an expression for the imaginary part of ρ_{21} using the equation (153.1).

As a final check, we can consider the case where the two lasers are on resonance $\Delta_p = \Delta_c = \Delta = 0$ and where the control Rabi frequency Ω_c dominates all the other rates. The equations (160) are then

$$\left\{ \begin{array}{l} \rho_{33} \approx \left(1 + \frac{\Gamma_{12}}{2\gamma_{13}} \right) \rho_{22} \\ 1 - \left(1 - \frac{\Gamma_{23}}{2\gamma_{13}} \right) \rho_{33} \approx \frac{\Omega_c^2}{\Omega_p^2} \left(\frac{\Gamma_{12}}{2\gamma_{13}} \right) \rho_{22} \end{array} \right. \quad (162)$$

These equations lead to the expression for the imaginary part of ρ_{12}

$$\mathcal{I}m(\rho_{12}) \approx 2 \frac{\gamma_{13} \Omega_p}{\Omega_c^2} \quad (163)$$

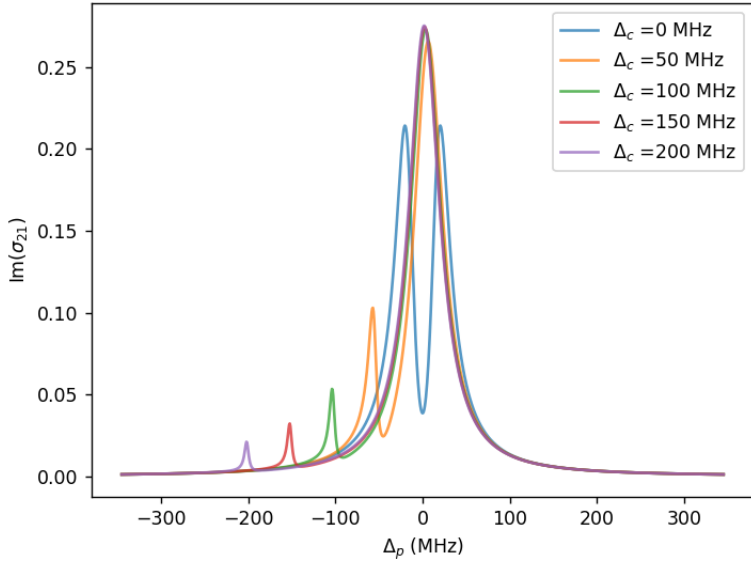
This is what we expect from the simple solution (152) derived in the previous subsection of this appendix.

A final remark is that to be even more precise, we can replace in the final expressions (161) of the excited states populations ρ_{22} and ρ_{33} the term Γ_{23} by $\Gamma_{23} + \Gamma_{13}$.³³ Thus, the equation (153.2) is the same as (149.2). The only remaining approximation to solve the optical Bloch equations (149) in the steady state regime is

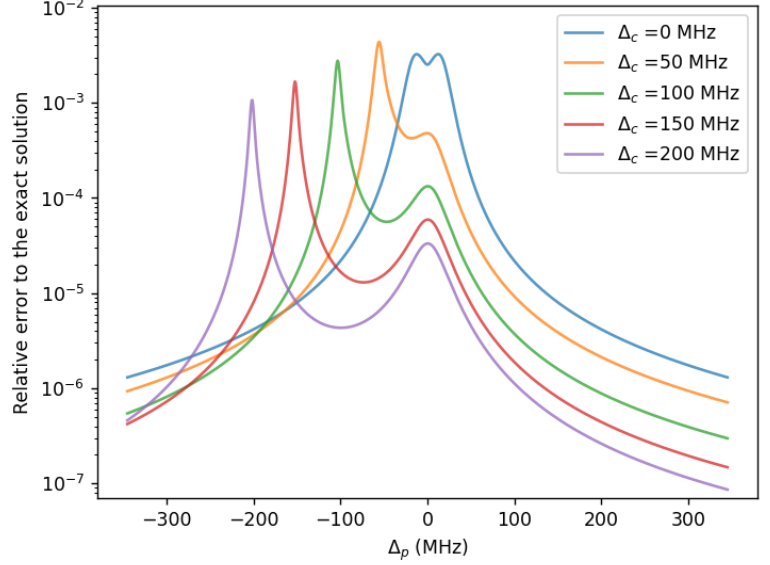
$$\Omega_p \mathcal{I}m(\rho_{12}) = \Gamma_{12} \rho_{22} + \underbrace{\Gamma_{13} \rho_{33}}_{\text{neglected}}. \quad (164)$$

Taking this last remark into account, we plot the expression of $\mathcal{I}m(\rho_{21})$ in Figure 37, where the transition linewidths Γ_{12} and Γ_{23} are those of the neutral calcium atom level that we consider in this work. The parameter Γ_{13} is chosen arbitrarily to be quite high compared to the estimate we made in subsection **3.1.2 Neutral calcium atoms** to test our approximation (164). The other parameters are typical parameters that we use in the experiments.

³³Though, be careful not to change it in the definition of γ_{23}



(a) Analytical expression of $\mathcal{I}m(\rho_{21})$



(b) Relative error of the analytical solution

Figure 37: (a) Analytical expression of $\mathcal{I}m(\rho_{21})$ and (b) its relative error compare to the exact one, for Rabi frequencies $(\Omega_p, \Omega_c) = (13.6, 37.4)$ MHz, transition linewidth $(\Gamma_{12}, \Gamma_{23}, \Gamma_{13}) = (34.5, 2.5, 0.35)$ MHz and laser linewidth $\gamma_p = \gamma_c = 1$ MHz

We see in Figure 37 (b) that for different values of the detuning of the control laser Δ_c , the relative error with respect to the exact solution is less than 1 percent. Moreover, it is maximal near the two-photon resonance when the population of level $|3\rangle$ is maximal, which is consistent with the approximation (164). Note that the relative error appears to be proportional to the ratio of the dipole decay rates, either the $4^1P_1 \leftrightarrow 4^1S_0$ transition or the $4^1D_2 \leftrightarrow 4^1P_1$ transition, and the quadrupole decay rate for the $4^1D_2 \leftrightarrow 4^1S_0$ transition. This intuition is confirmed because when we consider the typical value of Γ_{13} of a few tens of hertz estimated in (107), we take $\Gamma_{13} = 35$ Hz, and keeping all the parameters as in Figure 37, we get the same pattern for the relative error as on Figure 37 (b) but roughly multiplied by a factor 10^{-4} so that the maximum relative error is around 10^{-6} .

References

- [1] Andrew J. Daley, Immanuel Bloch, Christian Kokail, Stuart Flannigan, Natalie Pearson, Matthias Troyer, and Peter Zoller. Practical quantum advantage in quantum simulation. *Nature*, 607:667–676, July 2022.
- [2] I. M. Georgescu, S. Ashhab, and Franco Nori. Quantum simulation. *Rev. Mod. Phys.*, 86:153–185, Mar 2014.
- [3] Antoine Browaeys and Thierry Lahaye. Many-body physics with individually controlled rydberg atoms. *Nature Physics*, 16(2):132–142, jan 2020.
- [4] Kai-Niklas Schymik, Bruno Ximenez, Etienne Bloch, Davide Dreon, Adrien Signoles, Florence Nogrette, Daniel Barredo, Antoine Browaeys, and Thierry Lahaye. In situ equalization of single-atom loading in large-scale optical tweezer arrays. *Physical Review A*, 106(2), aug 2022.
- [5] Dolev Bluvstein, Harry Levine, Giulia Semeghini, Tout T. Wang, Sepehr Ebadi, Marcin Kalinowski, Alexander Keesling, Nishad Maskara, Hannes Pichler, Markus Greiner, Vladan Vuletić, and Mikhail D. Lukin. A quantum processor based on coherent transport of entangled atom arrays. *Nature*, 604(7906):451–456, apr 2022.
- [6] Andrea Muni, Léa Lachaud, Angelo Couto, Michel Poirier, Raul Celistrino Teixeira, Jean-Michel Raimond, Michel Brune, and Sébastien Gleyzes. Optical coherent manipulation of alkaline-earth circular rydberg states. *Nature Physics*, 18(5):502–505, mar 2022.
- [7] E. L. Raab, M. Prentiss, Alex Cable, Steven Chu, and D. E. Pritchard. Trapping of neutral sodium atoms with radiation pressure. *Phys. Rev. Lett.*, 59:2631–2634, Dec 1987.
- [8] Harold J. Metcalf and Peter van der Straten. *Optical Traps for Neutral Atoms*, pages 149–164. Springer New York, New York, NY, 1999.
- [9] T. E. Mehlstäubler, K. Moldenhauer, M. Riedmann, N. Rehbein, J. Friebe, E. M. Rasel, and W. Ertmer. Observation of sub-doppler temperatures in bosonic magnesium. *Phys. Rev. A*, 77:021402, Feb 2008.
- [10] R. Paschotta. Laser noise, 2023.01.09.
- [11] Antoine Browaeys Alain Aspect, Fabien Bretenaker and Claude Fabre. Quantum optics 1: Lasers, 2020.

- [12] M Squared Lasers Ltd. Solstis narrow linewidth, tunable cw ti:sapphire laser user manual v10.3, 2010.
- [13] Jacques Dupont-Roc Claude Cohen-Tannoudji and Gilbert Grynberg. *Lagrangian and Hamiltonian Approach to Electrodynamics, The Standard Lagrangian and the Coulomb Gauge*, chapter 2, pages 79–168. John Wiley & Sons, Ltd, 1997.
- [14] Jacques Dupont-Roc Claude Cohen-Tannoudji and Gilbert Grynberg. *Other Equivalent Formulations of Electrodynamics*, chapter 4, pages 253–359. John Wiley & Sons, Ltd, 1997.
- [15] Renato Renner. Lecture notes: Quantum information theory, 2019.
- [16] Frieder Lindenfelser. Broadband cooling on a forbidden transition in a novel high-optical-access ion trap, 2017.
- [17] Felix Vogt. Creation of cold and dense ensembles of calcium atoms, 2009.
- [18] Michael Fleischhauer, Atac Imamoglu, and Jonathan P. Marangos. Electromagnetically induced transparency: Optics in coherent media. *Rev. Mod. Phys.*, 77:633–673, Jul 2005.
- [19] Hilmar Oberst. Resonance fluorescence of single barium ions, 1999.
- [20] Christopher J Foot. *Atomic physics*. Oxford master series in atomic, optical, and laser physics. Oxford University Press, Oxford, 2007.
- [21] Wolfgang Demtröder. Laser spectroscopy 2: Experimental techniques, 2015.
- [22] Franck Laloë Claude Cohen-Tannoudji, Bernard Diu. Chapter xix - quantization of electromagnetic radiation. In Franck Laloë Claude Cohen-Tannoudji, Bernard Diu, editor, *Quantum Mechanics, Volume 3: Fermions, Bosons, Photons, Correlations, and Entanglement*. Wiley, second edition edition, 2019.
- [23] Gillenhaal Beck. Towards background-free readout and cooling in $^{40}\text{Ca}^+$ with a multiply-connected quadrupole transition, 2020.
- [24] C. Wieman and T. W. Hänsch. Doppler-free laser polarization spectroscopy. *Phys. Rev. Lett.*, 36:1170–1173, May 1976.

- [25] Ran Finkelstein, Samir Bali, Ofer Firstenberg, and Irina Novikova. A practical guide to electromagnetically induced transparency in atomic vapor, 2022.
- [26] L. Giner, L. Veissier, B. Sparkes, A. S. Sheremet, A. Nicolas, O. S. Mishina, M. Scherman, S. Burks, I. Shomroni, D. V. Kupriyanov, P. K. Lam, E. Giacobino, and J. Laurat. Experimental investigation of the transition between autler-townes splitting and electromagnetically-induced-transparency models. *Physical Review A*, 87(1), jan 2013.
- [27] Jacques Dupont-Roc Claude Cohen-Tannoudji and Gilbert Grynberg. *The Dressed Atom Approach*, chapter 6, pages 407–514. John Wiley & Sons, Ltd, 1998.
- [28] Luís E.E. de Araujo, Silvânia A. Carvalho, Luciano S. Cruz, A.A. Soares, Armando Mirage, Daniel Pereira, and Flavio C. Cruz. Optogalvanic detection of velocity-selective optical pumping in an open, cascade atomic medium. *Optics Communications*, 281(4):626–632, 2008.
- [29] Michael A. Nielsen and Isaac L. Chuang. *Quantum Computation and Quantum Information: 10th Anniversary Edition*. Cambridge University Press, 2010.
- [30] J.R. Johansson, P.D. Nation, and Franco Nori. Qutip 2: A python framework for the dynamics of open quantum systems. *Computer Physics Communications*, 184(4):1234–1240, 2013.
- [31] K. Brodie and S. Neate. Features and operation of hollow cathode lamps and deuterium lamps, 2022.12.14.
- [32] Laser sensor using the opto-galvanic effect highly stable output, see-through cathode optimality for laser wavelength calibration and laser frequency stabilization, 2023.01.09.
- [33] R. L. Cavasso-Filho, A. Mirage, A. Scalabrin, D. Pereira, and F. C. Cruz. Laser spectroscopy of calcium in hollow-cathode discharges. *J. Opt. Soc. Am. B*, 18(12):1922–1927, Dec 2001.
- [34] Michael Horbatsch and Marko Horbatsch. Classical versus quantum calculation of radiative electric quadrupole transition rates for hydrogenic states. *Canadian Journal of Physics*, 100(10):429–436, oct 2022.
- [35] R. Paschotta. Faraday isolators, 2023.02.21.

- [36] R. Paschotta. Etalons, 2023.02.13.
- [37] HighFinesse. Wavelength meter ws6-200 series, 2022.
- [38] Martin Adam Sepiol. Master thesis: Frequency stabilization of a 729 nm diode laser to an external high finesse reference cavity, 2012.
- [39] IntraAction Corp. Model aom-af1 series acousto-optic modulator / frequency shifter.
- [40] Ludwig Erasmus de Clercq. Transport quantum logic gates for trapped ions, 2015.
- [41] M Squared Lasers. Locking the solstis to a reference, 2014.
- [42] A. Trautmann, M. J. Mark, P. Ilzhöfer, H. Edri, A. El Arrach, J. G. Maloberti, C. H. Greene, F. Robicheaux, and F. Ferlaino. Spectroscopy of rydberg states in erbium using electromagnetically induced transparency. *Phys. Rev. Res.*, 3:033165, Aug 2021.
- [43] NIST. Atomic spectra database.
- [44] David H. Meyer, Paul D. Kunz, and Neal Solmeyer. Nonlinear polarization spectroscopy of a rydberg state for laser stabilization. *Appl. Opt.*, 56(3):B92–B96, Jan 2017.
- [45] Daniel Manzano. A short introduction to the lindblad master equation. *AIP Advances*, 10(2):025106, feb 2020.
- [46] Mitchel Weissbluth. Chapter 1 - angular momentum. In Mitchel Weissbluth, editor, *Atoms and Molecules*, pages 1–47. Academic Press, 1978.
- [47] R. Clark Jones. A new calculus for the treatment of optical systems. description and discussion of the calculus. *J. Opt. Soc. Am.*, 31(7):488–493, Jul 1941.
- [48] Marc D. Levenson and Satoru S. Kano. Chapter 3 - saturation spectroscopy. In Marc D. Levenson and Satoru S. Kano, editors, *Introduction to Nonlinear Laser Spectroscopy (Second Edition)*, pages 78–129. Academic Press, second edition edition, 1988.
- [49] Stanford Research Systems. About lock-in amplifiers, 2022.12.14.
- [50] R. Paschotta. Acousto-optic modulators, 2022.12.14.
- [51] Jia-ming Liu. *Acousto-optic devices*, page 357–440. Cambridge University Press, 2005.

Declaration of originality

The signed declaration of originality is a component of every semester paper, Bachelor's thesis, Master's thesis and any other degree paper undertaken during the course of studies, including the respective electronic versions.

Lecturers may also require a declaration of originality for other written papers compiled for their courses.

I hereby confirm that I am the sole author of the written work here enclosed and that I have compiled it in my own words. Parts excepted are corrections of form and content by the supervisor.

Title of work (in block letters):

Laser frequency stabilization in a calcium optogalvanic lamp

Authored by (in block letters):

For papers written by groups the names of all authors are required.

Name(s):

Pagot

First name(s):

Louis

With my signature I confirm that

- I have committed none of the forms of plagiarism described in the '[Citation etiquette](#)' information sheet.
- I have documented all methods, data and processes truthfully.
- I have not manipulated any data.
- I have mentioned all persons who were significant facilitators of the work.

I am aware that the work may be screened electronically for plagiarism.

Place, date

Zürich, 12/03/2023

Signature(s)



For papers written by groups the names of all authors are required. Their signatures collectively guarantee the entire content of the written paper.