Master in Photonics

MASTER THESIS WORK

LASER FREQUENCY STABILIZATION TO EXCITED RYDBERG TRANSITIONS USING ELECTROMAGNETICALLY INDUCED TRANSPARENCY

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Laser frequency stabilization to excited Rydberg transitions using electromagnetically induced transparency

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Abstract.
Rydberg atoms with principal quantum number \( n \gg 1 \) have extraordinary atomic properties including tunable long range dipole-dipole interactions that lead to the so called Rydberg blockade. Atoms excited to these particular levels have been shown to be excellent candidates to implement several quantum information tasks, from two-qubit gates to single photon sources. In this work, we demonstrate Electromagnetic Induced Transparency (EIT) using Rydberg levels in a hot gas of rubidium atoms. We then use the EIT window as a reference to stabilize the frequency of the coupling laser.

Keywords: Rydberg atoms, Electromagnetic Induced Transparency, Quantum Information, Laser stabilization.

1. Introduction

1.1. Rydberg atoms

A Rydberg atom is an atom with one or more electrons excited to levels with very high principal quantum number \( n \gg 1 \). These atoms exhibit peculiar properties, including an exaggerated response to electric and magnetic fields, long decay times and electron wave functions that approximate, under some conditions, classical orbits of electrons around the nuclei [1]. Their energy structure can be described accurately by

\[
E_{nlj} = -\frac{R_y}{(n - \delta(n)lj)^2} = -\frac{R_y}{(n^*)^2}
\] (1)

Where \( R_y \) is the Rydberg constant and \( n^* = n - \delta(n)lj \), being \( \delta(n)lj \) the quantum defect [2], \( l \) the orbital quantum number and \( j \) the total angular momentum. Fundamental properties of Rydberg atoms scales with \( n \) [3], for example, the orbital radius \((\langle r \rangle \propto n^2)\), the dipole moment \((\mu \propto n^2)\), the radiative lifetime \((\tau \propto n^3)\) or the electrical polarizability \((\alpha \propto n^7)\). Hence, huge atomic properties can be achieved by high excited Rydberg atoms.

Two of the most interesting phenomena that make these atoms suitable for quantum information are their tunable long range interaction and the so called dipole blockade. Both phenomena arise from the electrical dipole-dipole interaction between the atoms. If we
consider a pair of atoms initially in the Rydberg state $|rr\rangle$ separated by a distance $R$, the dipole-dipole interaction potential for this system can be written in atomic unit as \[ V(R) = \frac{\hat{\mu}_1 \cdot \hat{\mu}_2}{R^3} - \frac{3(\hat{\mu}_1 \cdot R)(\hat{\mu}_2 \cdot R)}{R^5} \quad (2) \]

Where $\mu_{1,2}$ are the dipole moments operators associated with the two atoms. In the pair basis $\{|rr\rangle, |rr'\rangle, |rr''\rangle, |r'\rangle|r''\rangle \ldots \}$, using eq. 2 and just considering the coupling between the two states closest in energy $\langle rr|V(R)|r'r''\rangle$, the energy of the two atoms state $|rr\rangle$ is shifted by a quantity $\Delta E$, which depends on $R$ and $n$ \[4\] \[2\] (see fig. 1(a)(b)). This means that we have a tunable long range interaction that depends on the states of the atoms. That is an essential requirement to implement two-qubit quantum gates. As an example, the interaction strength of Rb atoms to the 100S Rydberg state is $10^{12}$ times strongly than the Rb ground state atoms \[4\].

The dipole blockade is a phenomenon that ensures that only one Rydberg excitation can be created within a certain radius from the excited atom, which is called Blockade radius ($R_b$). For the case of a pair of atoms resonantly excited from $|g\rangle$ to $|r\rangle$ at Rabi frequency $\Omega$, the atoms will populate the state $|rr\rangle$ at a rate $\Omega$. If the atoms are close together, the dipole-dipole interaction will cause the detuning of the state $|rr\rangle$ with respect to the laser resonance, preventing the excitation of the $|rr\rangle$ state. The blockade mechanism enables the deterministic creation of a single excitation distributed among a cloud of $N$ atoms within $R_b$ (see fig. 1 (c)) and it can be used to implement quantum gates \[5\] \[7\].

1.2. Electromagnetic Induced Transparency (EIT)

EIT is a coherent phenomenon arising in a three-level system coupled by a weak probe field and a strong coupling field. The presence of this strong field modifies the optical response of the probe beam (see fig. 2). On resonance, the coherently driven medium experiments normal dispersion and, depending on the system and the particular values of the different parameters, it can becomes completely transparent for the probe field. The steepness of the dispersion can be tuned with the coupling power. This phenomenon is widely used to obtain ultra slow light, longitudinal pulse compression or even light storage \[6\]. The linear response of a medium is related to its energy level-structure and determined by the first order electrical susceptibility $\chi^{(1)}$. 

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**Figure 1.** Rydberg dipole-dipole interaction. (a) Transformation from atomic to pair state basis. (b) Dipole-dipole interaction shift the energy of the state $|rr\rangle$ by $\Delta E$. The state $|rr\rangle$ is detuned respect the laser excitation, hence the state $|rr\rangle$ cannot be populated. (c) Within the $R_b$ radius just only one Rydberg excitation is allowed.
The EIT phenomenon can be illustrated as a consequence of a quantum pathway interference [8]. Once an intense beam is coupling the levels \(|2\rangle - |3\rangle\) (see fig.2), the absorption from \(|1\rangle\) to \(|3\rangle\) can occur following two different ways. Directly \(|1\rangle - |3\rangle\) or indirectly \(|1\rangle - |3\rangle - |2\rangle - |3\rangle\). Because of the coupling beam is much more intense than the probe, the two possible paths have a probability amplitude of the same order of magnitude and thus a destructive interference leading to EIT becomes possible.

To achieve EIT involving Rydberg states a ladder scheme has to be used. The essential features of EIT can be quantitatively described by a semi-classical approach. Within the dipole approximation and considering the rotating wave approximation, the Hamiltonian of a ladder three-level system (see fig.2(b)) interacting with a coupling laser with real Rabi frequencies. \(\Delta = \omega_c - \omega_p\) and \(\Delta_p = \omega_p - \omega_{21}\) are the field detunings with respect atomic resonances, \(\Gamma_i\) are the corresponding level lifetimes. \(\chi^{(1)}\) as a function of \(\omega_p\) relative to the atomic resonance frequency \(\omega_{21}\), for a radiatively broadened two-level system with radiative width \((\gamma_{31})\) (dashed line) and a EIT system with resonant coupling field (solid line). (c) \(\text{Re}[\chi^{(1)}]\) where the dispersion coefficient is defined as \(\beta = \frac{1}{2}\text{Re}[\chi^{(1)}]\). (c)/(d) taken from [8].

\[
\hat{H} = -\frac{\hbar}{2} \begin{pmatrix} 0 & \Omega_p & 0 \\ \Omega_p & -\Delta_p & \Omega_c \\ 0 & \Omega_c & - (\Delta_p + \Delta_c) \end{pmatrix}
\]

The optical response of the probe field \((E_p)\) is determined by \(\chi^{(1)}(\omega_p)\). In the weak probe regime \((\Omega_c >> \Omega_p)\), the polarization is \(P \approx \frac{1}{2} \epsilon_0 E_p \chi^{(1)}(\omega_p)e^{-i\omega_p t} + c.c.\) = \(-\mu_{21}N \rho_{21} e^{-i\omega p t} + c.c.\) where \(N\) is the density of dipoles. Solving eq.4 in the steady state regime, considering the weak probe approximation and a Doppler broadened medium \((\delta \nu = \vec{k} \cdot \vec{v})\) in counter propagation configuration \((\vec{k}_p \cdot \vec{k}_c = -\frac{(2\pi)^2}{\lambda_c^2}\) the contribution to the total electrical susceptibility due to the atoms moving at velocity \(v\) can be obtained following [10] by

\[
\chi(v)dv = \frac{i \mu_{21}^2 / \epsilon_0 \hbar}{\gamma_{21} + i \Delta_1 - i \omega_p v} \frac{\Omega_p^2 / 4}{\gamma_{31} - i(\Delta_1 + \Delta_2) - i(\omega_p - \omega_c) v} N(v)dv
\]
Where $\gamma_{ij}$ correspond to the associated decay rates and are given by $\gamma_{ij} = (\Gamma_i + \Gamma_j)/2$, $\Gamma_i$ being the natural decay rate of the state $|i\rangle$. The total susceptibility is obtained by integrating eq.5 over the velocity distribution $N(v)$, which is conventionally taken Maxwellian (1D along the direction of the lasers) for atomic Doppler broadened gases [10].

The first EIT features involving highly excited Rydberg states were observed in [11]. Due to the interference nature of the EIT phenomenon sub-natural linewidth features are observed. These properties make EIT spectra suitable for spectroscopic purposes. In particular, EIT spectra involving Rydberg states can be used to coherently detect Rydberg states [11], measure absolute transition frequencies of high Rydberg states [12] or even use this signal as a reference to stabilize a laser to high excited Rydberg states [13]. Recently EIT has been used to make a single photon source [14] and it is used in a photon-photon gate proposal [4], this last topic being the long term goal of our experiment.

1.3. Objectives of the work

The objective of this work is to demonstrate laser frequency stabilization to highly excited Rydberg states using EIT spectroscopy on a rubidium (Rb) thermal vapour cell. Our first goal will be to build the experimental setup. After that we will do a complete characterization of our system in order to understand its behaviour, and thus be able to choose the optimal parameters that will give the best signal for our purpose. The last step will be to stabilize the laser frequency and to characterize its performance.

2. Experimental methods

2.1. Setup/Physical system

For all our experiments we use a Rb vapour cell of length ($L$) 75mm (Thorlabs GC25075-RB). Rb is commonly used in quantum optics experiments because its level structure allows laser cooling, it posses several long lived ground states and also because the first excited state is easily accessible with diode lasers (780nm). Moreover Rb is a metal alkaline and hence suitable to be excited to high Rydberg levels. In our case the $^{87}Rb$ isotope has been used. The relevant physical properties are described in [15].

The experimental setup is shown in fig.3. For the probe beam ($\lambda_p = 780nm$) we use light amplified by a semiconductor taper amplifier (Toptica BOOSTA) coming from another experiment and produced by a commercial diode laser (Toptica TA Pro). The frequency of the probe can be locked on an atomic transition thanks to saturation spectroscopy technique using an extra Rb cell at room temperature. The coupling beam ($\lambda_p = 473 - 485nm$) is produced by a commercial frequency doubled diode laser system (Toptica TA-SHG Pro), whose wavelength is tuned using a commercial wave meter to the desired Rydberg level (see fig.4). The values for the transition wavelengths are taken from [12].
Both beams pass through double passage AOMs to adjust their frequencies. The probe laser is resonant with the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 2)$ transition. The coupling and probe beams are counterpropagating through the Rb cell and they have a waist of 200$\mu$m and 400$\mu$m respectively. The transmission of the probe beam through the cell is monitored by an avalanche photodiode (APD) (Hamamatsu C5460), while the coupling laser frequency is scanned over the $5P_{3/2} \rightarrow \text{Rydberg}$ transition. Fig.4(a) shows the energy level structure of the system.

The Rb cell is placed inside a $\mu$-metal shield to reduce the effect of stray electromagnetic fields. Copper wires warped around the aluminum tube shown in fig. 3 are used to control the gas temperature by means of Joule effect. We use bifilar wiring in order not to create a magnetic field on the atoms as the current is circulating in the heater. Moreover four strip electrodes made of copper foils are attached to the surface of the cell, allowing the generation of tunable electric fields inside the cell. This configuration ensures full control of the system variables.

To compare the experimental data with the theoretical predictions, the model described by eq.5 is used. This model reproduces with very good agreement the experimental data when we are in the weak probe regime and the transition $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 2)$ is not saturated ($I_{\text{probe}} < I_{\text{sat}}$). The fit of the experimental transmission with the theoretical expression $I/I_0 = e^{-\alpha L}$ (where $I_0$ and $I$ are the input and the output powers respectively, while $\alpha$ is the absorption coefficient as derived in [10]), allow us to extract information otherwise non accessible experimentally (e.g. $\Omega_c, \gamma_{31}$ and $N$). All the contributions to $\alpha$ from the different hyperfine lines need to be included weighting their contribution with respect to their relative dipole moment $\langle 5S_{1/2}(F) | \hat{\mu} | 5P_{3/2}(F') \rangle$ and relative degeneracies.

In fig.4(b) we show a typical EIT spectrum obtained by scanning the coupling laser across the $5P_{3/2} \rightarrow nS_{1/2}$ resonance while the probe laser is kept locked on the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 2)$ transition.

Fig.5 shows the prediction of the model described by eq.5 in comparison to the experimental data for two different situations, with and without the magnetic shield. The only fit parameters are $\Omega_c(nL_j) = \frac{\langle 5P_{3/2} | \hat{\mu} | nL_j \rangle \cdot E}{\hbar}$, $\gamma_{31}(nL_j) = \gamma_{31}(nL_j \rightarrow 5P_{3/2})$ and $N$. $N$ can be related to the temperature ($T$) (see [15]). Note that in fig.5(b) the fitted function mimics better the function calculated with the theoretical value of $\gamma_{31}$ than in fig.5(a), proving that...
stray electromagnetic fields make the EIT peaks considerably lower and broader.

Figure 4. (a) Energy level diagram of the Rb$^{87}$ ladder system and the scheme of the dipole allowed two photon transitions to a highly Rydberg excited state. (b) Typical EIT spectrum. Three EIT resonances are observed because of the existence of different velocity classes of atoms. Due to the Doppler mismatch the hyperfine splitting of the $5P_{3/2}$ state scale with $\Delta \omega_c = (1 - \frac{\lambda_c}{\lambda_p}) \Delta \omega_p$ [11].

Figure 5. Experimental EIT spectrum across $nD$ transition (black) compared with the fitted theoretical model (red) and the same function with the theoretical value of $\gamma_{31}$ (blue) extracted from [4]. (a) Rydberg state 25$D_{3/2}$, Rb cell at room temperature without magnetic shield, $\Omega_c(25D_{3/2}) = 3.178 MHz$, $\gamma_{31}(25D_{3/2}) = 3.345 MHz$. (b) Rydberg state 50$D$, Rb cell at $T = 30^\circ C$ with magnetic shield, $\Omega_c(50D_{5/2}) = 1.95 MHz$, $\gamma_{31}(50D_{5/2}) = 1.271 MHz$, $\gamma_{31}(50D_{3/2}) = 1.274 MHz$.

2.2. Parameter study of Rydberg EIT

In this section different sets of experimental data are shown and analysed with the aim of characterizing the influence of the different parameters on the EIT spectra for our system. The response of the EIT spectra to the variation of the coupling power $P_c$, the probe power $P_p$ and the $T$ are analysed. The spectra are recorded scanning the coupling laser across resonance, while maintaining the probe laser locked.

Keeping the probe power constant, in the weak probe regime, we observe that within the range of powers used in the experiment the height of the EIT peaks grow linearly with $P_c$. On the other hand, within the powers used in the experiment, no significant variation of the FWHM of the peaks have been observed. Typically the FWHM observed are around 4MHz, narrower than the natural linewidth of the intermediate state ($\Gamma_{5P_{3/2}} = 6 MHz$). In this regime, the different values of $\Omega_c$, $\gamma_{31}$, and $N$ can be extracted fitting the experimental data with the formula derived in [10]. As an example, the set of values of $\Omega_c(nD_{5/2})$ and $\gamma_{31}(nD_{5/2})$ extracted using this method for different $P_c$ are shown in fig.6. Fig.6(a) shows the extracted $\Omega_c$ as a function of $P_c$. The experimental points are in good agreement with a square root
fit \( \Omega_c \propto \sqrt{P_c} \). This shows the validity of this method to extract otherwise non accessible parameters like \( \Omega_c \) or \( \hat{\mu}_{ij} \). Fig.6(b) shows that the experimental value of \( \gamma_{31}(nD_{5/2}) \) is not in agreement with the theoretical value, but higher. This could be explained by considering that this model does not take into account extra dephasing terms like for example the collisional broadening.

The theoretical model described in eq.5 predicts the existence of an optimal temperature to achieve the highest peaks of transparency when \( P_c \) and \( P_p \) are fixed. A temperature change induces a variation of the number of Rb atoms present in the vapour as well as their velocity distribution. The experimental results illustrating how the temperature influence the EIT spectra are shown in fig. 7. For this Rydberg level we observe that the optimal T is around 310K.

\( P_p \) is not included in the model used in the previous analysis. For this reason, the behaviour of the EIT spectra was analysed as a function of the \( P_p \) just experimentally. We observed that is observed that the height of the peaks increase with \( P_p \). On the other hand the FWHM increases slightly with \( P_p \). As we will see in the next section both behaviours will play in our favour. Also apparent in fig.8(a) is that the line shape of the EIT feature shows enhanced absorption just below and above the two-photon resonance. This effect arises from the wavelength mismatch between the coupling and probe lasers [11] and vanishes when \( P_p \) is increased (see fig.8(b)).

**Figure 6.** Rydberg state: 50D, T=30°C, \( P_p = 482nW \) (a) Values of \( \Omega_c(50D_{5/2}) \) extracted from the fit of the experimental data (blue) compared with a fit \( \Omega_c \propto \sqrt{P_c} \) (red) (b) Values of \( \gamma_{31}(50D_{5/2}) \) (blue) compared with the theoretical extracted from [4](green). Mean of the experimental values (red).

**Figure 7.** Peak Height [\( \Delta \) Transmission] relative to the transition \( 5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3) \rightarrow 40D_{5/2} \). Experimental values (blue), theoretical prediction (red). Thermodinamical properties of Rb taken from [15].
2.3. Laser frequency stabilization

We now want to use an EIT peak as a reference to stabilize the frequency of the coupling laser. We act on the current of the diode laser to lock its frequency to the maximum of the transmission spectrum. In order to obtain an Error Signal (ES) to feedback on the control current, we use the frequency modulation spectroscopy technique [16]. By using this technique an ES proportional to the derivative of the reference signal can be obtained (odd function). Using this type of ES, to maintain the frequency stabilized to the desired value, our system will have to keep the ES around 0. In order to obtain larger ESs, we increase $\Omega_c$ by reducing the coupling beam waist down to 200 $\mu$m. The used setup is sketched in fig.9 and examples of ES’s are shown in fig.10(a).

From the previous section it can be concluded that, in order to obtain a higher spectroscopic signal and consequently a better ES, high $P_c$ and $P_p$ are needed. But, as we want these stabilized lasers to be used for other experiments, ideally we should use the lowest possible power. Because of the power needed for the coupling is much bigger than for the probe, we will be limited by $P_c$.

As the dipole elements $\langle 5P_{3/2}(F)|\hat{\mu}|nD \rangle$ and $\langle 5P_{3/2}(F)|\hat{\mu}|nS \rangle$ scale with $1/n^{3/2}$ [2] the power needed to keep the same EIT signal scales with $n^3$. It means that to obtain equivalent Signal to Noise Ratio (SNR) the $P_c$ required increases significantly as we increase $n$. Moreover the dipole matrix elements for the 5p-ns transitions are an order of magnitude less than 5p-nd.

To evaluate the performance of the lock, we compute the SNR considering as a signal the amplitude of the ES, and as a noise the rms value of the ES out of resonance. The combined linewidth of the coupling and and probe lasers has been also estimated. To do that, the rms noise of the locking signal when the laser is locked was measured and divided by the gradient of the unlocked ES.

In fig.10(b) an example of a lock performance analysis as a function of the $P_c$ can be seen. It is noticeable that the SNR and the estimated linewidth saturates up to certain $P_c$. It shows that up to certain values of $P_c$ the lock stability is no longer limited by the ES gradient (and hence by the coupling laser power used in the locking scheme) and starts to be limited by probe laser intensity fluctuations or electronic noise.
Laser frequency stabilization to excited Rydberg transitions using EIT

Figure 9. Schematic setup for the laser stabilization. The probe beam is kept locked on $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 2)$ transition and is modulated at $110\text{KHz}$ using an AOM. The ES it is obtained by mixing the signal coming from the APD with the reference signal using a lock in amplifier. Then this is sent to a PID microcontroller that generates the control signal. Finally the control signal will feed the diode current of the coupling laser.

Figure 10. Lock performance study for the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3) \rightarrow 40D_{5/2}$ transition. $P_p = 2.15\mu\text{W}$ (a) Error signals obtained with different $P_c$ (smoothed over 20 points) (b) SNR and estimated laser linewidth

3. Conclusions

We have demonstrated laser frequency stabilization to excited states transitions using cascade EIT involving highly excited Rydberg states using a thermal vapour cell of Rb. As the technique is based on atomic coherences, regardless of being in a Doppler broadened medium, one can obtain resonance widths which are even smaller than the natural linewidth of the intermediate states. This allows us to obtain steep error signals to feedback the lasers and thus to obtain narrow laser linewidths. Furthermore, the signal used in this system is an absolute two photon transition reference.

In this work a laser stabilization with a good SNR (SNR=70) with relatively low $P_c$ (100mW) up to a 79D Rydberg state has been reported. This prove the validity of this method as a lock technique when stabilization to highly Rydberg states is required. The main advantage of this method is its experimental simplicity. Some modifications on the setup are required to lock onto higher Rydberg states. A better error signal could be obtained by reducing the coupling beam waist to increase the $(\Omega_c)$ and by incrementing the modulation frequency $(f)$ to reduce the electronic noise $(N_e)$ since $N_e \propto 1/f$. The next step of the experiment is to build a cold Rydberg setup using this system to stabilize the coupling laser.
Moreover, recently interesting experiments involving hot Rydberg atoms have been carried out [17] [18], and new and interesting experiments could be done using thermal vapour cells. Since we have demonstrated that the variables of our system can be very well controlled and tuned, it opens the possibility to use also our setup for a wide range of new experiments in the quantum optics field.

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