LASER PHYSICS AND SPECTROSCOPY IN ALKALI VAPOR-NOBLE GAS MIXTURES

BY

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DISSERTATION

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Abstract

Experiments are described in which the lower excited states of alkali-rare gas diatomic molecules are probed in alkali vapor-noble gas mixtures by laser spectroscopy. A two color scheme for pumping the alkali D$_2$ line laser has been demonstrated that offers a quantum efficiency above one. More than 100 cm$^{-1}$ of thermal energy is extracted from the gain medium for every laser photon. An optical amplifier based on free$\rightarrow$free transition of alkali-rare gas pairs has been demonstrated in the 793.3-794.3 nm region of the spectrum. Lasing on specific hyperfine atomic transitions—and, as a result, a 50% D$_2$ line laser efficiency increase—has been demonstrated when alkali vapor-noble gas mixtures are pumped with a circularly polarized light in contrast to a classical pumping scheme (linearly polarized light).
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# Table of Contents

Chapter 1  INTRODUCTION .................................................. 1

Chapter 2  THEORETICAL BACKGROUND ................................. 3
  2.1  Alkalis and Alkali Atomic Lasers ................................. 3
  2.2  Atomic Lasers Pumped by the Photoassociation of Atomic Pairs  4
  2.3  Collapse of a Three Level Laser Into a Two Level System ....... 6
  2.4  Selection Rules ..................................................... 7
  2.5  Circular Polarization of Light and Spin Exchange in Alkali-  
       Noble Gas Mixtures ............................................... 9

Chapter 3  INVESTIGATION OF ENERGY ROUTING MECHANISMS  
          BETWEEN STATES OF ALKALI-NOBLE GAS MOLECULES  
          AND ALKALI ATOMS ............................................. 12
  3.1  General Description of the Experimental Arrangement .......... 13
      3.1.1  Energy Stabilization Assembly ............................. 13
      3.1.2  Alkali-Noble Gas Cells ................................... 15
  3.2  Investigation of the Minimum Energy Separation For a Three 
       Level Laser ..................................................... 16
  3.3  Two Color Pumping ................................................ 19
      3.3.1  Realization of a Laser with a Quantum Efficiency Above 
             One ......................................................... 19
      3.3.2  Realization of RbXe Molecular Amplifier Based on 
             Free→Free Transitions of Rb-Xe Collision Pairs .......... 28
      3.3.3  Observation of Quantum Oscillations on the D_2 Line 
             Red Satellite of RbXe Pairs ............................... 33
      3.3.4  Detection of Stimulated Raman Scattering by Two Color 
             Excitation .................................................. 36
  3.4  Circularly Polarized Pump and Hyperfine Molecular and Atomic 
       Structure ....................................................... 39
      3.4.1  ASE Experiments ............................................ 39
      3.4.2  Circularly Polarized Alkali Lasers (CPALs) ............... 45
      3.4.3  Observation of Quantum Oscillations on the D_2 Line 
             Red Satellite of K-Noble Gas Pairs ....................... 50
Chapter 1

INTRODUCTION

Alkali atoms have been central to atomic, molecular, and laser physics since the early development of quantum theory, primarily because of their structure. Having one valence electron allows for cross-sections and excited state structures to be calculated with a high degree of precision. The simplicity of alkali atomic systems is contrasted by the small number of alkali atoms and consequent lack of diversity in their physical properties. The addition of noble gases to alkali vapors greatly enriches the variety of available physical systems without compromising the simplicity of a single valence electron configuration. Interatomic interactions of such mixtures have been studied since the 1970s [1, 2]. Noble gases and alkalis form weakly-bound diatomic molecules in an excited state and, because their ground states are dissociative, these species are referred to as excimers (excited dimers). Because of their filled valence shells, the noble gas atoms have a high-lying first excited state (∼8.44 eV for Xe) and, as a result, act as a hard sphere while interacting with alkali atoms. Several groups reported experiments probing the lowest $^2\Pi$ states [3] and ab initio calculations predicted the interaction potentials for states as high as $^9\Pi$ [4]. Furthermore, Happer and co-workers [5] and Eden et al. [6] discovered alkali-rare gas excited states, correlated with $n^2S_{\frac{1}{2}}$, $(n - 2)^2D + Rg$ in the separated atom limit, that are optically connected to ground.

In 2008 Readle et al. [7] demonstrated the first alkali laser pumped via the blue satellite associated with an alkali D$_2$ line perturbed by a specific rare gas species that generated laser emission on the alkali D$_1$ and D$_2$ lines simultaneously. This laser was dubbed an Exciplex Pumped Alkali Laser (XPAL). These laser systems have extraordinary quantum efficiencies (> 95% for Rb and > 98% for Cs, for example) and a relatively broad pump acceptance spectrum (> 1 nm or 0.5 THz) owing to the spectral width of the alkali blue satellites. However, pumping into an alkali-rare gas blue satellite suffers from a low absorption coefficient (reduced absorption coefficient for Cs-Xe is ∼ 2 × 10$^{-36}$ cm$^{-5}$, for example). Therefore, this process has not yet found extensive practical application. Nevertheless, this
discovery has introduced several powerful new techniques such as photoexcitation spectroscopy, opening new horizons in studying the physics of alkali atoms and alkali-noble gas molecules, as well as fundamental physics, in general.

This dissertation describes the result of a series of experiments designed to probe in detail the spectroscopy and energy flow in the lowest electronic excited states of the Cs, Rb, and K - rare gas diatomic molecules. One and two color pumping of these systems discussed in Section 3.3 has enabled the efficiency of lasers operating on the alkali D$_2$ line to be improved considerably, and oscillators having a quantum efficiency above unity have been realized. Furthermore, the presence of a weak energy barrier (heights of 2-10 cm$^{-1}$) has been detected in two color laser spectroscopic experiments for the first time. Such barriers have been predicted on the basis of theory [8] but have not been observed previously. These energy barriers have been implemented in this work for the first demonstration of light amplification utilizing free $\rightarrow$ free molecular transitions.

The subject of Section 3.2 is the study of minimum energy separation between two upper laser states. This effect was previously investigated by Hewitt et al. [9] in 2012 with a different approach from this work and alkali-noble gas pair; however a confirmation of this result was required to claim its generality. The minimum energy separation for RbXe gas mixture has been measured to be $\sim$1 kT which has a good agreement with the value of $\sim$0.7 kT obtained by Hewitt et al. In addition to experimental data, this work provides theoretical calculations in which the gradual reduction of laser efficiency is simulated at the point where the minimum energy separation limit is reached. In short, the minimum energy separation, $\Delta E$, is the value below which the thermal coupling between two laser states becomes non-negligible, and eventually transforms the three or four level system into a two level system that is incapable of lasing. This is a fundamental physical effect which causes a significant reduction of laser efficiency when the value of $\Delta E$ becomes comparable to kT.

The third major thrust of this work is the demonstration of an optical excitation of alkali atoms via the alkali-noble gas molecular transitions with circularly polarized light (Section 3.4). The proposed pumping mechanism results in stimulated optical emission on alkali atomic transitions associated with specific hyperfine alkali states. This grants a 50% increase of the atomic laser efficiency and a significant decrease in the lasing threshold value.
Chapter 2

THEORETICAL BACKGROUND

2.1 Alkalis and Alkali Atomic Lasers

All alkali atoms (Li, Na, K, Rb, Cs) have a similar energy level structure. Figure 2.1 shows a partial energy level diagram of the Rb atom. The first two excited states \( n^2P_{\frac{1}{2}} \) and \( n^2P_{\frac{3}{2}} \), where \( n \) is the principal quantum number and \( n = 2, 3, 4, 5, 6 \) for Li, Na, K, Rb, and Cs, respectively), as well as the ground state \( n^2S_{\frac{1}{2}} \), are of primary interest in this work and are known as resonance states because

![Figure 2.1: Partial energy level diagram of atomic Rb.](image)
they are optically connected to ground. Transitions between ground and the first doublet states of the atom are known as the D₁ and D₂ lines.

From this point on, when the alkalis or their properties are mentioned, it will be supposed that the alkali vapor (T > 350 K) is referred to.

The first two excited states of the alkalis have lifetimes ranging from 15 to 35 ns [10]. The lifetime of the \( n^2P_{\frac{1}{2}} \) is typically several ns shorter than the \( n^2P_{\frac{3}{2}} \) state.

It has been recognized by Krupke et al. [11] that the alkali vapor may be efficiently pumped by a high power, narrow-line laser diode array on the \( n^2P_{\frac{3}{2}} \leftarrow n^2S_{\frac{1}{2}} \) transition followed by fast collisional relaxation to the \( n^2P_{\frac{1}{2}} \) state. This results in a population inversion and lasing on the alkali D₁ line (\( n^2P_{\frac{1}{2}} \rightarrow n^2S_{\frac{1}{2}} \)). The quantum efficiency of such a laser is >95% for Cs and >98% for other alkalis.

This particular gas laser is called a diode pumped alkali laser (DPAL). The main advantage of the system is the ability to convert the laser diode output, having a low quality beam profile (\( M^2 \approx 100 \)), into a laser beam of high (\( M^2 \approx 1 \)) quality, which is common for gas lasers. Laser diodes are known for their low price and high overall efficiency (>70%); in combination with the high quantum efficiency of the alkali lasers and the fact that multiple laser diodes may be used to pump the alkali vapor, alkali lasers became great candidates for the scalable high power lasers.

The primary disadvantage of an optically pumped atomic laser system is the narrow pump acceptance spectrum (typical values are up to several GHz [12]), implying that the pump source must match the alkali D₂ line to a high precision. This, unfortunately, does not allow the use of inexpensive laser diodes without temperature and wavelength stabilizing options to pump DPALs. Several other processes such as thermal lensing and soot formation (because of the photo-dissociation of organic gases used for faster collisional relaxation) also become problems for this type of the laser.

2.2 Atomic Lasers Pumped by the Photoassociation of Atomic Pairs

In an effort to overcome a narrow acceptance spectrum, associated with the atomic lasers, another pumping scheme for the alkali lasers was proposed by Readle et al. in 2008 [7]. It was suggested to use broad (>500 GHz) continua associated
Figure 2.2: Energy diagram demonstrating the operational principle of the RbXe XPAL.

with free—free transitions of alkali-noble gas molecules to pump alkali atoms to their $n^2P_{\frac{3}{2}}$ states, as shown in Fig. 2.2, for the RbXe molecule. In this process, alkali-noble gas molecules are optically excited from the ground state to the dissociative $B^2\Sigma_{\frac{1}{2}}^+$ state. Molecules pumped into this state dissociate in 10-100 ps, leaving the alkali atomic fragment in the $5^2P_{\frac{3}{2}}$ state. Such transitions in alkalis were thoroughly studied in the past [1, 3, 2]. The spectra associated with these transitions are known as blue satellites.

The pumping mechanism described above inverts the populations on both D$_1$ and D$_2$ line alkali transitions, compared to only D$_1$ for the previously described system, allowing lasing to be achieved on both alkali D lines. This feature becomes important in the case of sodium XPALs, having a potential to be used as a pump source for sodium guide stars [13]. Lasing on both Na D lines was previously demonstrated by Hewitt et al. [14] in 2012.

Having a much broader pump acceptance spectrum than the atomic alkali laser,
the laser pumped via the photoassociation of atomic pairs exhibits an absorption
coefficient in the alkali blue satellite that is \( \sim 4 \) orders of magnitude smaller than
that at the \( \text{D}_2 \) line itself. This serves as the main factor limiting the practical use
of these laser systems.

Although the alkali lasers pumped by photoassociation did not find their ap-
plication as industrial or military lasers due to the weak pump absorption, their
unique excitation mechanism allows one to use these system to study a variety of
alkali (and alkali-noble gas) properties as well as fundamental physical problems.

2.3 Collapse of a Three Level Laser Into a Two Level
System

The fundamental laser physics problem of the three level system collapse harks
back to the time when lasers had not yet been practically demonstrated. The
classic paper by Schawlow and Townes [15], along with the detailed discussion
of the theory of the optical MASER system, mentioned that the energy separation
between two upper energy states (\( \Delta E \)), shown in Fig. 2.3, must be much greater
than \( kT \) (where \( k \) is Boltzmann’s constant and \( T \) is the system’s temperature in
Kelvin).

The reason for this restriction (\( \Delta E \gg kT \)) is the increasingly strong thermal

\[
\begin{align*}
|0\rangle & \\
|1\rangle & \\
|2\rangle & \\
\end{align*}
\]

\( \Delta E \gg kT \)

Figure 2.3: A conventional three level laser system. The green arrow represents
the relaxation process of the system from state \( |2\rangle \) to \( |1\rangle \), and \( \Delta E \) is the energy
difference between states \( |2\rangle \) and \( |1\rangle \).
coupling between $|2\rangle$ and $|1\rangle$ that occurs as the value of $\Delta E$ approaches $kT$. Assuming that the Maxwell-Boltzmann distribution is valid for the current system, the population distribution between the two states at equilibrium is governed by the equation:

$$N_2 = N_1 \exp\left(-\frac{\Delta E}{kT}\right),$$

(2.1)

where $N_1$ and $N_2$ are the populations of $|1\rangle$ and $|2\rangle$, respectively. However, it is questionable if Eq. 2.1 accurately describes the actual population distribution for gaseous gain media during the process of lasing, because it is an expression that assumes thermal equilibrium whereas the population of the energy levels undoubtedly changes quickly with time.

In the limit $\frac{\Delta E}{kT} \to 0$, the populations of both closely-spaced energy levels become equal. In this case, a three level laser system essentially collapses into a two level system which cannot be used as a gain medium (when optically pumped).

The actual value of $\frac{\Delta E}{kT}$ for which the collisional coupling between $|1\rangle$ and $|2\rangle$ begins to significantly erode the efficiency of the laser was studied by Hewitt et al. [9] in 2012. In his experiments, monitoring the efficiency of the D$_2$ laser while photopumping Cs-Kr-Ar trios in the ground state found the critical value of $\frac{\Delta E}{kT}$ to be 0.7. Hewitt’s experiments were the first to explore this question, and additional experiments are desirable to confirm the results.

### 2.4 Selection Rules

Before introducing the selection rules for atomic spectra, it is useful to briefly review Russell-Saunders (or LS) coupling, which labels atomic states (Fig. 2.1) as:

$$^{2S+1}L_J$$

where $S$ is the total spin quantum number, $L$ is the total orbital angular momentum quantum number, and $J$ is the total angular momentum quantum number. The values of the total orbital angular momentum quantum number $L = 0, 1, 2, 3, 4, \ldots$ are assigned the letters S, P, D, F, G, ..., respectively.

Selection rules under the electric dipole approximation state that single photon
transitions between electronic states in atoms must obey the following rules:

\[ \Delta L = \pm 1 \]  \hspace{1cm} (2.2)
\[ \Delta S = 0 \]  \hspace{1cm} (2.3)
\[ \Delta J = 0; \pm 1 \]  \hspace{1cm} (2.4)

where \( \Delta L, \Delta S, \Delta J \) are the differences of the corresponding values between the initial and final atomic states. Equation 2.4 is not valid for transitions from a state with \( J = 0 \) to another state with \( J = 0 \) for which \( \Delta J = \pm 1 \) only. Those rules may be intuitively understood remembering that they represent the principle of the conservation of the angular momentum.

Photons carry an angular momentum equal to \( \hbar \) [16]. Therefore, when absorbing or emitting one photon, the atomic system should change its orbital angular momentum by the same value (\( \pm 1 \)). This implies that \( ^2S \rightleftharpoons ^2P, \ ^2P \rightleftharpoons ^2D, \ ^2D \rightleftharpoons ^2F \), etc., transitions are allowed but \( ^2S \rightleftharpoons ^2S, \ ^2P \rightleftharpoons ^2P, \ ^2S \rightleftharpoons ^2D \), etc., transitions are forbidden.

The same rules hold for two photon transitions, but the polarization of the light must be taken into account. Two forms of circular polarization (clockwise and counterclockwise) represent two eigenstates comprising all other possible polarizations. They represent photons with positive and negative angular momenta. Therefore, the circularly polarized wave carries only +1 or -1 values of the angular momentum, in contrast to randomly polarized radiation, where both values are present, and a linearly polarized wave (both +1 and -1 values are present, as well), which can be written as a combination of two opposite circular polarizations:

\[ \mathbf{E} = \hat{x}|E_0|e^{-j\beta z} = \left( \frac{|E_0|}{2} + j\hat{y}\frac{|E_0|}{2} \right) e^{-j\beta z} + \left( \frac{|E_0|}{2} - j\hat{y}\frac{|E_0|}{2} \right) e^{-j\beta z}, \]  \hspace{1cm} (2.5)

where \( \hat{x} \) and \( \hat{y} \) are orthogonal unit vectors in the plane transverse to the direction of propagation of the wave, \( E_0 \) is the amplitude of the wave, \( \beta \) is the wave vector, and \( z \) is the longitudinal spatial coordinate.

Based on the considerations above, the angular momentum selection rule for the two photon processes stimulated by the linearly or randomly polarized light become:

\[ \Delta L = 0, \pm 2, \]  \hspace{1cm} (2.6)
For the circularly polarized light, the selection rule reduces to:

$$\Delta L = \pm 2.$$ (2.7)

Equation 2.7 implies that $^2S \leftrightarrow ^2S$, $^2P \leftrightarrow ^2P$, $^2D \leftrightarrow ^2D$, etc., two photon atomic transitions may be suppressed by using circularly polarized light.

This property of circularly polarized light will be used later in this work to suppress the $7^2S_{\frac{1}{2}} \leftrightarrow 5^2S_{\frac{1}{2}}$ two photon transition in atomic Rb.

It should be pointed out that the selection rules are valid for atomic transitions; however, they may be violated in the case of the molecular transitions where all the values of the angular momentum are projected onto the axis of a molecule.

### 2.5 Circular Polarization of Light and Spin Exchange in Alkali-Noble Gas Mixtures

Polarized noble gas atoms (especially $^{129}$Xe) are known for their ability to produce strong magnetic resonance (MR) signals, and, accordingly, they are currently of considerable value for both studies of fundamental physics and medicine [17]. This section will be using the term polarized Xe, but it should be kept in mind that only certain noble gas isotopes are able to produce a magnetic resonance signal. A discussion of the noble gases used for producing magnetic resonance signals goes beyond the scope of this work, but can be found in [17].

Nuclear spin polarization, $P$, is usually described by a simple formula:

$$P = \frac{N^+ - N^-}{N^+ + N^-},$$ (2.8)

where $N^+$ and $N^-$ are the populations of the ground and excited spin states of an atom, respectively. Small difference between the energy levels associated with the spin states leads to a small polarization of atoms at room temperatures. Thus, a technique allowing for the production and storage of hyperpolarized (non-equilibrium distribution of population in nuclear spin states) Xe gas was required to efficiently use it in magnetic resonance imaging.

It has been shown in [18] that the noble gas nuclei can be efficiently polarized by a spin exchange process using alkali-noble gas mixtures (500 Torr of the noble gas was used) with addition of 1 Torr of $N_2$ (RbHe, RbNe, RbKr, and RbXe were
used in this experiment). This process implies that the weak coupling between the electron spin of the alkali-metal atom, the rotational angular momentum of the molecule, and the nuclear spin of the noble-gas atom in a magnetic field cause the exchange of the electron (alkali) and nuclear (noble gas) spin due to a relatively long lifetime of the alkali-noble gas molecules. The polarization of the Rb atoms, in turn, was achieved by pumping the alkali-noble gas cells with circularly polarized light, propagating along the applied magnetic field. The parameters of this process were further studied and improved [19, 20, 21] but the basic concept has remained unchanged.

The mechanism for optical pumping of alkali vapor utilized, for the purpose of creating the hyperpolarized noble gas atoms, is shown in Fig. 2.4. Circularly polarized light at the wavelength corresponding to the alkali D$_1$ line transition (it has been shown in [20] that D$_2$ line transitions may also be used) pumps a cell in the direction of an applied magnetic field. The cell is maintained at 320-450 K and filled with the alkali vapor, 100-2000 Torr of noble gas, and 1-20 Torr of N$_2$. Due to the presence of the magnetic field, the selection rules allow only $\Delta m = \pm 1$ transitions ($m$ is the magnetic quantum number). Thus, the circularly polarized light excites the Rb atoms to only one of their hyperfine $^2P_{\frac{1}{2}}$ states ($m = +1/2$ in the case of $\sigma^+$ photoexcitation). Surprisingly, collisions do not introduce significant mixing between the $+1/2$ and $-1/2$ spin states of the alkalis [19]. Reference [22]

Figure 2.4: Optical pumping of Rb hyperfine levels with circularly polarized light, $\sigma^+$ means that the light is left-hand circularly polarized.
provides a clear introduction to the selection rules associated with the atoms in the magnetic field. The presence of the N$_2$ gas quenches the radiative relaxation of the excited +1/2 spin state of the alkali and stimulates a non-radiative collisional relaxation, having a rate of 1:2 for +1/2 $\rightarrow$ −1/2 and +1/2 $\rightarrow$ +1/2 spin transitions, respectively. Therefore, after some period of time, the system resides in the ground, spin +1/2 state, which is inaccessible to $\sigma^+$ light, and this gas mixture becomes transparent to $\sigma^+$ polarized light.

Quenching of the radiative $^2P_{\frac{1}{2}} \rightarrow ^2S_{\frac{1}{2}}$ transitions is important because the spontaneously emitted photons, emitted in random directions, are reabsorbed by the alkali vapor and almost completely scramble the polarization of the alkali atoms.

After the alkali vapor is polarized, the electron spin of the alkali atoms is slowly (typically minutes, depending on the pressure of the noble gas and the temperature of the cell) transferred to the nuclear spin of the noble gas atoms via the spin exchange process.

Produced in this way, hyperpolarized Xe atoms remain polarized for tens of minutes to hours, depending on the pressure of the noble gas and the magnetic field strength. It is worth mentioning that the relaxation process of the noble-gas nuclear spins is not caused by binary collisions in the gas mixture. This process occurs primarily through the interaction of the atoms with the walls of the cell [19].

This and previous sections briefly described the influence of circularly polarized light on the selection rules for atomic transitions. New physical phenomena are introduced, which do not appear in the case of the linearly or randomly polarized pumping. These concepts will become important in Section 3.4, which describes the pumping of the alkali-noble gas mixtures with circularly polarized light.
Chapter 3

INVESTIGATION OF ENERGY ROUTING MECHANISMS BETWEEN STATES OF ALKALI-NOBLE GAS MOLECULES AND ALKALI ATOMS

Alkali-noble gas mixtures grant a unique opportunity to investigate a variety of energy transfer mechanisms between the molecular and atomic states of alkalis. This work is focused on experimental exploration and investigation of these mechanisms with different types of spectroscopic techniques, the most important of which is photoexcitation spectroscopy.

All experiments described in this work were performed using an experimental arrangement comprising one or two Sirah PrecisionScan dye lasers with a linewidth of 0.05 cm$^{-1}$. The dye lasers were pumped with the second harmonic of a single Spectra-Physics Quanta-Ray Pro-350 Nd:YAG laser in order to avoid time jitter between pulses. The Nd:YAG laser operated at the repetition rate of 10 Hz. The dye lasers acted as pump or probe sources for various types of alkali-noble gas mixtures. The first section of this chapter explains the operational principles underlying specific aspects of experimental setups such as the energy stabilization assembly, alkali-noble gas cells, and the temperature controlled oven. Further sections describe experimental arrangements used in each individual experiment.

This chapter is arranged in such a way that each following section relies on the experimental results and conclusions obtained from the previous one. The first experiment on measuring the minimum energy separation between two upper laser levels in RbXe (Section 3.2) was performed in order to confirm results published by Hewitt et al. (Ref. [9]) for the CsAr gas mixture. The results of this section were used to build a conceptual model of the two color pumping mechanism in alkali-noble gas mixtures, discussed in Section 3.3. This concept implies utilizing the thermal energy of the system to excite upper laser states and, therefore, achieve a quantum efficiency (optical-to-optical) above one. Promising experimental results on utilizing free $\rightarrow$ free molecular transitions for the amplification of laser radiation (first demonstration of a free $\rightarrow$ free molecular amplifier) have been obtained utilizing a similar pumping scheme. Additional experiments which allow undulations on the red side of the Rb D$_2$ line to be investigated, and stim-
ulated Raman scattering (SRS) via two color photoexcitation spectroscopy to be explored, are also described in this section. Section 3.4 introduces a novel pumping mechanism for alkali-noble gas mixtures using circularly polarized light to selectively pump and achieve stimulated emission on certain hyperfine transitions of alkalis. This technique opens new horizons for the spectroscopy of alkali-noble gas molecules and allows for a significant increase in the performance of atomic lasers pumped by the photoassociation of atomic pairs.

3.1 General Description of the Experimental Arrangement

3.1.1 Energy Stabilization Assembly

Energy fluctuations of the second harmonic (532 nm) of the Nd:YAG laser used to pump the dye lasers are specified to be below 4%. However, energy fluctuations of the dye lasers pumped with the second harmonic of Nd:YAG varied in the range of 10-15% due to many reasons, most of which are related to the flow conditions of the laser dye solution. Such big fluctuations in the pump power result in poor signal-to-noise ratios, especially in the case of photoexcitation spectra measurements, which imply that the measured signal has a nonlinear dependence on the pump laser’s energy.

In order to decrease the described fluctuations, the energy stabilization assembly shown in Fig. 3.1 was placed after each dye laser. The pump energies were controlled using a polarizing beam splitter (PBS), a calibrated high-sensitivity pyroelectric detector, and an achromatic half-wave plate mounted in a motorized,
precision rotation mount as shown in Fig. 3.1. A LabView script with a feedback loop has been written for the current experiments in order to control and record the pump energies of the dye lasers.

The initial output beam from a dye laser had a vertical polarization. The polarization vector was precisely controlled (> 0.3 degree accuracy) by a rotational stage with a mounted achromatic half-wave plate. The remaining vertical polarization component (after the wave plate) was reflected by the PBS and dumped in a beam block. Horizontally polarized light passed through the PBS and was directed to the experimental setup. A beam sampler placed after a PBS picked 1-5% of the vertically polarized beam and steered it to the pyroelectric detector. The LabView script measured the energy of each individual pulse and compared this value with the desired pulse energy. If the measured energy was in a given range, the script recorded all the values of the other energy detectors used in the setup. In the case when more than ten energy pulses were out of the desired range, the motorized rotational mount changed the angle of the half wave plate in order to compensate for the difference between the actual and intended energy values.

The setup described here allows for accurate tuning of the pump energy while keeping the beamshape unchanged at the same time. In contrast, changing the energy of the pump laser (Nd:YAG) directly leads unavoidably to distortion of the dye laser’s beam profile.

It is important that the cross correlation function between the pyroelectric detector shown in Fig. 3.1 and other detectors placed downstream in the same beam path be regularly measured in order to assure that all the detectors pick the same laser pulse every time. The correlation function should always peak at zero; otherwise, the energy stabilization setup does not provide any benefit insofar as data quality is concerned.

Another reason why the stabilization assembly is required is a bell-shaped laser dye spectral profile. Wide-range scans (> 1 nm) introduce a smooth output energy change due to the dependence of the dye gain on the required wavelength. This effect may be completely compensated by the assembly described here, providing a real, flat (constant pump energy) excitation spectrum measurement.

In the future, the energy stabilization assembly will not be shown explicitly for the sake of diagram simplicity.
3.1.2 Alkali-Noble Gas Cells

Sealed quartz cells 2.5 cm in diameter and 10 cm in length, filled with alkali metal and noble gas, served as gain media in all experiments described below. The cell’s windows were tilted at an 11° angle in order to avoid undesirable Fabry-Perot effects. Alkali cells were placed into an oven driven by a proportional-integral-derivative (PID) temperature controller, enabling the cell’s temperature to be maintained within ±1 K, and were heated to the temperatures corresponding to the desired alkali number densities. The temperature dependence of the alkali number density, used for conversion, is shown in Fig. 3.2 for Na, K, Rb, and Cs.

A copper rod 5 mm in diameter and 12 cm in length was brought into contact with a side of the quartz cell inside the oven. The second end of the rod was fixed outside the cell, contacting ambient air at the room temperature. This cold finger was used in order to create the coldest point of the cell and, therefore, indirectly monitor the number density of the alkali of interest by measuring the temperature of the induced coldest point.

![Figure 3.2: Number densities of Na, K, Rb, and Cs as a function of temperature. Dependencies taken from [23].](image)
3.2 Investigation of the Minimum Energy Separation For a Three Level Laser

Alkali atomic lasers pumped at free→free molecular transitions give a rare opportunity to study the minimum energy separation $\Delta E$ (briefly discussed in Section 2.3) between levels 2 and 3 in a three-level laser system. For instance, the energy separation between the blue satellite of RbXe molecule and Rb D$_2$ line is 337 cm$^{-1}$. This value of $\Delta E$ corresponds to a temperature of 485 K (212 °C) which may be easily achieved. Therefore, the minimum value for $\Delta E$ may be estimated by investigating the dynamics of the atomic laser output intensity as a function of the cell’s temperature (Rb number density).

Previous studies of the minimum energy separation described in [9, 24] were performed by measuring the decay constant of the red side of the Cs-Ar-Kr excitation spectrum and plotting it versus the temperature of the cell. In the current experiment, an alternative approach (described below) was developed. A Rb-Xe gas mixture having a Xe number density of $1.8 \times 10^{19}$ cm$^{-3}$ (500 Torr) was chosen for this experiment. The reason for varying the alkali metal (from Cs to Rb) was to show that the value of the minimum energy separation between two states in a three level system is not a property of the specific laser system but rather is a universal value attributable to all alkali or, even more generally, all atomic and molecular gas lasers.

An experimental setup for measuring the minimum value of $\Delta E$ in RbXe is given in Fig. 3.3. A single dye laser, operating on Styril 8 (LDS-751) dye solution, was used to pump the Rb-Xe gas cell which was situated in a temperature controlled oven. The temperature of the oven was varied from 150 to 250 °C which corresponds to Rb vapor number densities from $10^{14}$ to $5 \times 10^{15}$ cm$^{-3}$ and the value of $\frac{\Delta E}{kT}$ from 1.15 to 0.93. An L-shaped optical cavity, built around the oven, was designed for horizontal polarization of the Rb D$_2$ line laser while the pump laser beam had vertical polarization and pumped the cell longitudinally. The initial (incident) and unabsorbed pump energies were measured with pyroelectric energy detectors 1 and 2 respectively. The unabsorbed power was measured in order to ensure that only a small portion (< 10%) of the pump was absorbed by the RbXe gas mixture and, therefore, the gain medium was pumped uniformly along the cell. Energy detector 3 measured the output energy of the Rb D$_2$ line laser. A narrow bandpass filter with the a bandwidth of 2 nm FWHM and a central wavelength of 780 nm (Rb D$_2$ line) was placed between the output coupler and
Figure 3.3: Experimental setup for determining the minimum energy separation, $\Delta E$, between the upper laser states of the RbXe XPAL.

The energy detector to block the pump laser’s residuals.

The wavelength of the dye laser was scanned over the entire breadth of the Rb-Xe blue satellite (758-763 nm). The output intensity of the Rb-Xe D$_2$ line laser (780 nm) was measured as a function of the pump wavelength while maintaining constant pump pulse energy. Known as a laser excitation spectrum, the dependence of the Rb-Xe laser pulse energy on pumping wavelength is shown in Fig. 3.4 for four different temperatures of the Rb-Xe cell. The output energy reduction at 760 nm emerges due to a two photon absorption process, and to a four wave mixing process in Rb, and will be discussed in detail in Section 3.4. These processes are not related to the study of the minimum energy separation and will be ignored in this section.

The plot in Fig. 3.5 shows the Rb D$_2$ line laser’s output energy dependence as a function of the cell’s temperature, expressed in eV. The vertical dashed line in Fig. 3.5 denotes the temperature corresponding to the energy separation $\Delta E$ between the pumped and upper laser levels. In other words, it shows the temperature at which the ratio $\frac{\Delta E}{kT}$ is equal to one. Note that the D$_2$ line laser’s intensity initially increases with T since its gain is proportional to the Rb atom number density, which grows nonlinearly with temperature (see the upper vertical axis in

17
Fig. 3.4: Excitation spectrum of Rb-Xe gas mixture at four different temperatures.

Fig. 3.5). However, at ~ 41 meV the output energy reaches a maximum value and then falls, approaching zero at 45 meV. This breakpoint, located at $\frac{\Delta E}{kT} \approx 1$ (or slightly above), indicates the beginning of the collapse of the two upper laser states. The two energy levels become indistinguishable because of the strong thermal coupling between them; therefore, this system collapses into a two level system which is not able to lase.

This experiment was performed at three different values of pump energy to ensure that the effect is not related to the depletion of the pump and, as a result, a non-uniform gain distribution along the cell. As can be seen from Fig. 3.5, the output power of the RbXe XPAL drops with an increase in temperature for all three values of the pump laser energy, indicating that the system is weakening because level 2 and 3 are, in effect, moving closer to each other.

The measured value of $\frac{\Delta E}{kT} \approx 1.1$ is in good agreement with the value of ~ 0.7, determined by a different methodology and alkali-noble gas mixture in [9]. These two experiments allow for a reasonably good approximation for a minimum energy separation between two upper laser levels after which the laser’s efficiency quickly drops.
Figure 3.5: Dependence of Rb-Xe D\textsubscript{2} line laser output power as a function of temperature (expressed in eV) and Rb number density.

The results obtained in this experiment were used to simulate the process of the energy level collapse with a time dependent rate equation model which is described in Chapter 4.

3.3 Two Color Pumping

3.3.1 Realization of a Laser with a Quantum Efficiency Above One

The experimental results discussed in the previous section have important implications for atomic and laser physics and I suggest several conclusions. The fact that the energy separation $\Delta E$ between levels 1 and 2 (Fig. 2.3) may be as small as
1 kT and still support laser emission raises the question of whether it is possible to pump a laser via an energy level that lies below the upper laser level (as shown in Fig. 3.6). Such a laser system (if successful) would have a value of quantum efficiency $\eta = \frac{A_{\text{pump}}}{A_{\text{Laser}}}$ above one; therefore, each emitted photon would extract a small portion of thermal energy, $\Delta E$, from the system, resulting in its cooling. Such a system would ameliorate any problems associated with gain medium cooling, slow heat transfer, thermal lensing, etc. In other words, it would eliminate factors that currently limit the scaling of high power laser systems. The process depicted in Fig. 3.6 has been of interest for almost two decades to the segment of the laser community that is pursuing internally cooled lasers [25].

In order to make a system with $\eta > 1$, the following pumping mechanism was proposed. The first step is equalization of the populations of state $|0\rangle$ and $|2\rangle$ (not shown in Fig. 3.6). This may be achieved in several different ways. It will be assumed that an optical pump is used to satisfy this requirement. The second (main) pump pulse (blue arrow in Fig. 3.6), corresponding to the $|0\rangle \rightarrow |1\rangle$ transition, is introduced shortly after the first pulse. It is important that the population of state $|1\rangle$, $N_1$, should be significantly smaller than $N_0$ and $N_2$ when the main pump pulse arrives. The energy transfer mechanism between states $|1\rangle$

![Figure 3.6: Hypothetical three level laser system with quantum efficiency above one. The curve to the right of the diagram is the Maxwell-Boltzmann distribution for state $|1\rangle$ at $\Delta E = 1kT$. The dashed vertical line denotes collisional coupling between the pumped and lasing states $|2\rangle$ and $|3\rangle.$](image-url)
and \( |2\rangle \) and the lasing conditions are described in detail for RbXe gas mixtures later in this section.

The concept suggested by Fig. 3.6 has been confirmed experimentally in two color pumping experiments and the results obtained have been published in *Applied Physics Letters* [26].

Figure 3.7 illustrates in detail the process responsible for the two color pumped atomic laser. The overall pumping process is divided into two stages, the first of which is similar to the approach introduced in Section 3.2. The Rb-Xe gas mixture was pumped at a wavelength of the Rb D\(_2\) line blue satellite. The only difference between this and the previous case is that the energy of the pump was fixed at a value which was slightly less than the lasing threshold. Therefore, when the Rb-Xe gas mixture was pumped with the first laser (\( \lambda_1 = 760 \text{ nm} \)) only, no lasing on the Rb D\(_2\) line was observed.

![Figure 3.7: Interatomic potentials for several of the lowest-lying electronic states of the diatomic molecule RbXe. Photoassociation of ground state Rb-Xe collision pairs at two wavelengths is indicated and, for illustrative purposes, the height of the weak potential barrier on the RbXe (\( A^2\Pi_{1/2} \)) potential has been exaggerated.](image)

Figure 3.7: Interatomic potentials for several of the lowest-lying electronic states of the diatomic molecule RbXe. Photoassociation of ground state Rb-Xe collision pairs at two wavelengths is indicated and, for illustrative purposes, the height of the weak potential barrier on the RbXe (\( A^2\Pi_{1/2} \)) potential has been exaggerated.
The second stage is the introduction of the primary pump pulse ($\lambda_2 = 793-797$ nm) on the red side of the D$_2$ transition. It arrived at an adjustable time delay ($\Delta t$) following the first pump pulse and drove the $A^2\Pi_{\frac{1}{2}} \leftarrow X^2\Sigma^+_{\frac{1}{2}}$ molecular transition. The purpose of this pump was to create a significant population in the $A^2\Pi_{\frac{1}{2}}$ state, a portion of which would dissociate to produce Rb ($5^2P_{\frac{1}{2}}$) atoms. Excitation of the $B^2\Sigma^+_{\frac{1}{2}}$ molecular state by the first pump pulse leads to populating the Rb ($5^2P_{\frac{1}{2}}$) state because the $B$ state is repulsive (dissociative). Therefore, the first blue pump is responsible for driving the D$_2$ line to near transparency, meaning that:

$$N_2 - N_0 g_2 \approx 0,$$

where $N_0$ and $N_2$ are the number densities of the respective states, and $g_0$ and $g_2$ are the degeneracies of the $5^2S_{\frac{1}{2}}$ and $5^2P_{\frac{1}{2}}$ atomic states of Rb, respectively.

Thus, the second pump reduces the population of the ground state ($N_0$) even more, resulting in a population inversion on Rb D$_2$ line transition, which leads to lasing or the generation of amplified spontaneous emission (ASE), in the absence of a resonator (in this experiment, the resonator was removed and the ASE signal was measured by energy detector 3; see Fig. 3.8).

The first and second (prime) pump laser beams, emitted by the two dye lasers, are shown in Fig. 3.8. Represented with red and blue, respectively, the first and second pump pulse beams are introduced by the Nd:YAG laser system that drives the two dye lasers. The dye lasers generated 8 ns pulses, and the time delay between them was set to 9 ns. The pulse shapes of the first and second pump

![Figure 3.8: Schematic diagram of the experimental arrangement. The acronym PBS refers to a polarizing beamsplitter.](image-url)
pulses, as well as the output ASE at 780 nm (Rb D\textsubscript{2} line), are shown in Fig. 3.9. It can be seen that the ASE is triggered by the second pump pulse and is narrower than this pulse. This is behavior typical of an optically pumped laser, and the delay time between the beginning of the λ\textsubscript{2} pulse and the onset of ASE is obvious.

As mentioned above, the second pump laser (A\textsuperscript{2}Π\textsubscript{1/2} ← X\textsuperscript{2}Σ\textsuperscript{+}\textsubscript{1/2}) populates the A\textsuperscript{2}Π\textsubscript{1/2} state and depletes the ground (5\textsuperscript{2}S\textsubscript{1/2}) state population. This process induces a population inversion between the 5\textsuperscript{2}P\textsubscript{3/2} and 5\textsuperscript{2}S\textsubscript{1/2} states and automatically triggers lasing on the Rb D\textsubscript{2} line. The D\textsubscript{2} line stimulated emission quickly depletes the 5\textsuperscript{2}P\textsubscript{3/2} state and pulls the population of the 5\textsuperscript{2}P\textsubscript{1/2} state up to the 5\textsuperscript{2}P\textsubscript{3/2} by burning a hole in its thermal velocity distribution (Fig. 3.10). This process is critical to the performance of the laser and is dependent on the Rb (5\textsuperscript{2}P\textsubscript{1/2}) + Xe → Rb (5\textsuperscript{2}P\textsubscript{3/2}) + Xe spin-exchange process.

As the \textsuperscript{2}P\textsubscript{1/2} atoms having thermal energies > 237 cm\textsuperscript{-1} (spin-orbit splitting in the 5\textsuperscript{2}P manifold) are connected to \textsuperscript{2}P\textsubscript{3/2} atoms in collisions with background

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure3.9.png}
\caption{Normalized pump laser and ASE (D\textsubscript{2} line, 780 nm) waveforms observed for Rb-Xe mixtures when Δt = 9 ns, λ\textsubscript{1} = 759.95 nm, λ\textsubscript{2} = 794.15 nm, [Rb] = 9.2 × 10\textsuperscript{14} cm\textsuperscript{-3}, and [Xe] = 1.8 × 10\textsuperscript{19} cm\textsuperscript{-3}. The energies absorbed by the gain medium from the first and second pulses are E\textsubscript{1} = 84 \mu J and E\textsubscript{2} = 124 \mu J, respectively.}
\end{figure}
Figure 3.10: The first two excited states of Rb, illustrating the Maxwell-Boltzmann distribution associated with the $^5{}^2P_{1/2}$ atom thermal velocity. The dashed line represents a hole burned in the Maxwell-Boltzmann distribution because of stimulated emission on the $^2P_{3/2} \rightarrow$ ground transition. Blue arrows indicate the particle flux that opposes the formation of the hole.

Xe atoms, stimulated emission of the newly formed $^2P_{3/2}$ atoms has the result of burning a hole in the velocity distribution for the $^5{}^2P_{1/2}$ species. The presence of a void in the distribution will, as is well known for electron velocity distribution in semiconductors, produce a particle flux in velocity space that is proportional to $\frac{dN}{dE}$, the slope of the distribution in both sides of the hole. This process implies that for each atom upconverted by the $^5{}^2P_{1/2} \rightarrow ^5{}^2P_{3/2}$ process the system loses $\sim 102$ cm$^{-1}$ (12.6 meV) of its thermal energy and is, therefore, cooled.

The quantum efficiency, $\eta$, of such a system should be calculated taking into account the energies absorbed from both pump lasers. The most straightforward way of doing this is to sum the quantum efficiencies of each pump, weighted by the absorbed energy at each pump wavelength:

$$\eta = \frac{\lambda_1}{\lambda_{laser}} \frac{E_{abs1}}{E_{abs1} + E_{abs2}} + \frac{\lambda_2}{\lambda_{laser}} \frac{E_{abs2}}{E_{abs1} + E_{abs2}},$$

(3.2)

where $\lambda_{1,2}$ and $E_{abs1,2}$ are the wavelengths and the absorbed energies of the first and the second pump pulse, respectively, and $\lambda_{laser}$ is the wavelength of the Rb D$\_2$ line laser which is equal to 780 nm. The quantum efficiency of the system described in this experiment was measured to be 1.08 at $\lambda_1 = 760$ nm and $\lambda_2 = 794.3$ nm.
One may question Eq. 3.2 since a number of processes, unrelated to the increase of state $5^2P_2$ population, may cause $E_{abs2}$ to dominate over $E_{abs1}$ and, therefore, artificially raise $\eta$ above unity. In order to eliminate this possibility, optical-to-optical efficiency curves were measured for both single and two-color pumping and the results are shown in Fig. 3.11. The abscissa shows the total energy absorbed in the RbXe cell (i.e. $E_{abs1} + E_{abs2}$).

These experiments demonstrate that the efficiency of the two color pumped laser is almost double that of its single color counterpart. It must be emphasized that the absolute efficiency of 1.9% does not reflect accurately the efficiency for a CW-pumped system (which is known to be > 30 % for even a single color laser [27]) because of the absence of an optical cavity. Therefore, the graph is given to consider relative intensities of the single and two color pumped lasers in this experiment, which differ by more than a factor of 1.7 in favor of the two color pumping case. This example demonstrates that the second pump indeed affects

![Figure 3.11: (Red Circles): Dependence of $D_2$ line (780 nm) ASE on $E_1 + E_2$, the sum of the energies absorbed by the first and second pump pulses. For all measurements, $[\text{Rb}] = 9.2 \times 10^{14} \text{ cm}^{-3}$, $[\text{Xe}] = 1.8 \times 10^{19} \text{ cm}^{-3}$, $\lambda_1 = 759.95 \text{ nm}$, $\lambda_2 = 794.76 \text{ nm}$, and $E_1$ was fixed at 85 $\mu$J. The time delay $\Delta t$ was maintained at 8 ns. (Black Squares): Similar data but recorded when $E_2 = 0$ and the laser is pumped only at 760 nm (Rb-Xe blue satellite).](image-url)
the population of the $5^2P_3^0$ state, and the quantum efficiency of the current system is greater than one.

In addition to the efficiency measurements, laser excitation spectra for two color pumped RbXe, RbAr, and RbKr gas mixtures were acquired. The spectrum shown in Fig. 3.12 was recorded by fixing the energy of the first ($\lambda_1 = 760$ nm) and second pump lasers at 85 $\mu$J and 135 $\mu$J, respectively, and scanning the wavelength of the second laser around the Rb D$_1$ line (793-797 nm). It can be seen that the maximum D$_2$ laser output corresponds to $\lambda_2$ pumping on the blue side of the Rb D$_1$ line. This result may be explained by the presence of a small energy barrier on the $A^2\Pi_1$ state which is shown qualitatively in Fig. 3.7 at $\sim$ 5.5 Å. Theoretical calculations [8] predict such energy barriers for several alkali-noble gas combinations but not for Rb-Xe. Experimental studies of these activation

![Figure 3.12: Laser excitation spectrum for Rb-Xe at 473 K, recorded by monitoring the D$_2$ line ASE energy while scanning the second laser over the 793.5 – 796.6 nm region. For these experiments, [Rb] = 9.2 x 10$^{14}$ cm$^{-3}$, [Xe] = 1.8 x 10$^{19}$ cm$^{-3}$, $\Delta t$ was fixed at 9 ns, and $E_1$ and $E_2$ were maintained at 85 $\mu$J/pulse and 135 $\mu$J/pulse (at $\lambda_2 = 794.4$ nm), respectively.](image)
barriers are challenging because these energy barriers are small (maximum height of \( \sim 20 - 30 \) cm\(^{-1} \)). As an example, the RbHe energy barrier has been observed in the past [28] at low temperatures (tens of K) but no studies have been reported at temperatures approaching 300 K and beyond.

The current experiment shows that, in addition to the demonstration of a laser having a quantum efficiency above one, the two color pumping technique may be employed to study weak features of molecular potentials that are inaccessible by other means.

Similar excitation spectra were recorded for other Rb-noble gas mixtures and similar behavior was observed in Rb-Ar (Fig. 3.13) and Rb-Kr mixtures.

Similar two color experiment was conducted when the wavelength of the first laser (\( \lambda_1 \)) was set to 780 nm (Rb D\(_2\) line). The output power of the Rb D\(_2\) line ASE was reduced by more than one order of magnitude (output energies <100 nJ) implying that detected emission was stimulated by the ground state depletion only (no thermal excitation process was involved). This experiment demonstrates the

![Figure 3.13: Spectral data similar to that of Fig. 3.12 but obtained for Rb-Ar at four values of [Rb], corresponding to cell temperatures between 443 K and 503 K. For these measurements, [Ar] = 1.1 \times 10^{19} \text{ cm}^{-3}, E_1 = 90 \ \mu\text{J}, E_2 = 140 \ \mu\text{J} (at \lambda_2 = 794.4 \text{ nm}), and \Delta t = 9 \text{ ns.}](image-url)
significance of the fist pumping stage where the population distribution is prepared prior to the arrival of the main pump pulse. It is critical that the excitation via the D\(_2\) line transition cannot be used to populate the \(^2P_{\frac{3}{2}}\) state. Furthermore, no photons with energies corresponding to the D\(_2\) line transition must present in the cavity when the second pump pulse arrives.

The goal of this experiment was to demonstrate the feasibility of a system with quantum efficiency above unity. The concept of such a system was successfully demonstrated but further steps are required to bring it to life. The dependence of the output energy as a function of the time delay between pump pulses should be thoroughly studied in future experiments. It is expected that the efficiency gradually reduces with the increase of the time delay approaching zero for \(\Delta t > 15-20\) ns. Study of the system behavior as a function of the time delay should be followed by pumping alkali-noble gas mixtures with high power constant wave or quasi-constant wave laser through the energy barrier of the \(A^2\Pi\) state.

3.3.2 Realization of RbXe Molecular Amplifier Based on Free\(\rightarrow\)Free Transitions of Rb-Xe Collision Pairs

Two color pumping experiments reveal the presence of the small energy barrier on RbXe \(A^2\Pi\) molecular state. It has been demonstrated (Section 3.3.1) that pumping from the ground state directly onto this barrier is the most preferable and efficient way for the two color pumping process.

The same free\(\rightarrow\)free molecular transition can be utilized to achieve amplification of a seed laser tuned to the wavelength corresponding to this transition. The energy diagram for this process is shown in Fig. 3.14.

An experimental setup similar to the one used for the two color pumping experiment was utilized to demonstrate the first free\(\rightarrow\)free molecular amplifier and is given in Fig. 3.15. In this setup, the first laser (the wavelength was fixed at the peak of the RbXe blue satellite, \(\lambda_1 = 760\) nm) had a vertical polarization. It passed through the alkali-noble gas cell and was reflected back onto itself by a high reflecting mirror. Such a double-pass scheme enabled depositing more energy to the gain medium, enhancing the amplifier efficiency. The polarization of the second dye laser (red arrows in Fig. 3.15), used as a seed, was changed to horizontal, in contrast to the two color pumping case. Therefore, the seed laser was reflected by the second PBS, passed through the gain medium, and reflected
again by the first PBS. It was then guided to the diffraction grating and reflected by it into a pyroelectric energy meter (detector 3). A combination of the diffraction grating and an iris (Fig. 3.15) was used in order to filter out the pump laser beam (blue color) propagating in the same direction with the seed laser. This filtering scheme significantly reduced the signal (energy of the seed laser) measured by the energy meter; however, it enables completely eliminating the pump laser signal, in contrast to a bandpass filter. Energies of the first and second laser were fixed at 3 mJ/pulse and 100 µJ/pulse, respectively.

The transmission spectra (measured by detector 3) of the RbXe gas mixture with respect to the seed laser for several different temperatures are given in Fig. 3.16. The black line represents a cold scan, i.e. when the cell temperature was 300 K. The data demonstrates that in the temperature range \(\sim 433 \sim 453\) K, the energy of the seed laser exceeds the transmitted energy through the cold cell indicating the presence of the amplification effect. The difference between the amplified signal and cold transmittance was small (\(\sim 1 \sim 1.5\) µJ); however, the transmission spectra were reproduced over several heating/cooling cycles and the amplification

![Energy Diagram](image)

**Figure 3.14:** Energy diagram demonstrating the operational principle of the free \(\rightarrow\) free RbXe molecular amplifier. The height of the weak potential barrier on the RbXe \(A^2\Pi_{3/2}\) potential has been exaggerated for illustrative purposes.
phenomenon was always observed. In the future, an alternative filtering scheme is desirable in order to improve the signal-to-noise ratio.

Similar results, demonstrating amplification, were obtained when the system was pumped through the Rb D$_2$ line directly, instead of the RbXe blue satellite. Transmission spectra identical to those in Fig. 3.16 were measured. This result implies that the excitation mechanism does not affect the observed effect, in contrast to the original two color pumping experiment (Section 3.3.1).

The transmission spectra of the system have also been studied when the wavelength of first laser was tuned to the Rb D$_1$ line. In this experiment, no amplification was observed; i.e., the transmission of the gas mixture in the 433-453 K temperature range was below the cold transmission scan. This was expected as no stimulated emission should be observed in a two level system. This sanity check allows us to confirm the consistency of the experimental data.
Figure 3.16: Transmission spectra of the RbXe gas mixture (600 Torr of Xe) with respect to the seed laser, as a function of wavelength for several different temperatures. For all measurements the first laser was pumping the system at $\lambda_{pump} = 760$ nm with energy of 3 mJ/pulse.

The discovered gain effect was barely observable due to the pulsed nature of the pump and seed pulses. Along with the laser systems with quantum efficiency above unity, the demonstrated amplifier requires a high power, long pulse, pump laser to reach its potential.

Despite the weak gain, the recorded data allows us to extract useful information about the studied system. The amplification was observed in a narrow temperature range suggesting that the thermal distribution of the population of the $A^2\Pi_{\frac{1}{2}}$ state changes and inversion in the local Franck-Condon region (around 5.5 Å) disappears. This effect may be studied in the future by exciting the alkali-noble gas pairs from this energy barrier to higher lying atomic states.

Further, Fig. 3.17 shows the transmission spectrum of the RbXe gas mixture at $T = 443$. Red and blue curves represent the case when the pump laser was turned off and on, respectively. In order to estimate the height of the energy barrier, the
difference between the transmission spectra obtained without the pump (regular absorption) and with the pump (difference between blue and red curves in Fig. 3.17) was calculated and is shown in Fig. 3.18 in black. The photoexcitation spectrum of the RbXe gas mixture recorded in the two color pumping experiment is shown in the same plot with blue dots for comparison. The peaks of both spectra perfectly match, having a maximum at \(\sim 794.3\) nm. The peak of the black curve (Fig. 3.18) should not be confused with the maximum gain coefficient of the system which occurs on the left side of the peak (see Fig. 3.17). Thus, two different experiments independently demonstrate the presence of the energy barrier on the \(A^2\Pi\) molecular state.

In summary, experiments described in this section have demonstrated the first free\(\rightarrow\)free molecular transition amplifier. It worth mentioning that, classically,

![Figure 3.17: Transmission spectra of the RbXe gas mixture (500 Torr of Xe) with respect to the seed laser as a function of wavelength at T=443 K. Blue and red curves show transmittance of the gas cell with the presence and the absence of the pump laser (760 nm), respectively. Black curve represents transmittance of the cell at T = 300 K (cold scan).](image)
Figure 3.18: Black curve represents the difference of transmission coefficients of RbXe cell with and without pumping the blue satellite (red curve [Fig. 3.17] subtracted from blue curve). Blue dots represent the photoexcitation spectrum of the RbXe gas mixture recorded in the two color pumping experiment (Fig. 3.12).

molecular oscillators and amplifiers are realized on bound→free or bound→bound transitions.

3.3.3 Observation of Quantum Oscillations on the D₂ Line Red Satellite of RbXe Pairs

As mentioned before, the two color pumping of RbXe gas mixtures was implemented with the pump wavelengths \( \lambda_1 = 779 - 784 \text{ nm} \) and \( \lambda_2 = 792 - 797 \text{ nm} \) corresponding to the Rb D₂ and D₁ lines, respectively. While the use of these wavelengths does not support thermal excitation of atoms to higher states, several other interesting physical phenomena have been observed.

Figure 3.19 shows the photoexcitation spectrum obtained by fixing the wavelength of the second pump laser at 794.7 nm and scanning the wavelength of the
first laser from 779 nm to 784 nm. The effect of the first pump laser on the excitation spectrum due to the leakage of scattered photons through the bandpass filter (Fig. 3.8) was taken into account and, therefore, it represents a corrected photoexcitation spectrum.

Graphs given in Fig. 3.19 provide important information about the system. The decrease in energy of the Rb D$_2$ line ASE at $\lambda_1 \approx 780.1$ nm, most likely, occurs because of the radiation trapping of the photons in the vicinity of the Rb atomic resonance. Two other clearly distinguishable features are of particular interest. First, the excitation spectrum dramatically changes its slope with changing temperature. The most probable explanation for this phenomenon is the change of the absorption coefficient, which, in turn, reflects the population distribution for RbXe molecules in the $A^2\Pi_2$ state. Therefore, excitation spectra similar to those

![Figure 3.19: Laser excitation spectrum for Rb-Xe at four different temperatures, recorded by monitoring the D$_2$ line ASE energy while scanning the first laser over the 779 – 784 nm region. For these experiments, $\lambda_2 = 794.7$ nm, [Xe] = $1.8 \times 10^{19}$ cm$^{-3}$, $\Delta t$ was fixed at 8 ns, and $E_1$ and $E_2$ were maintained at 1.5 mJ/pulse and 450 $\mu$J/pulse, respectively.](image-url)
of Fig. 3.19 may be used to calculate the vibrational distribution of the population in the $A^2 \Pi_{\frac{1}{2}}$ and $A^2 \Pi_{\frac{3}{2}}$ states if a proper model and interaction potentials are used.

The most interesting feature can be observed at 463 K, where a well-pronounced oscillation pattern (black curve in Fig. 3.19) appears in the excitation spectrum. A high resolution scan (Fig. 3.20) taken at $T = 453$ K shows the oscillations in detail. The reproducibility of this effect was confirmed by changing the gas cell’s positions, and repeating multiple heating/cooling cycles. The frequency of the observed undulations ($\sim 30.9$ GHz) does not change with wavelength, suggesting that this effect is not related to the oscillations of the ground state or excited state wavefunctions associated with the RbXe molecules. The oscillations were initially attributed to Fabry-Perot effect arising from an optical element. However, absence of any optics having optical thickness of 9.75 mm, and the fact that the oscillations may be observed only in a certain temperature range and only on the red side of the Rb $D_2$ line, indicate that the effect is likely caused by an interfer-

![Figure 3.20: High resolution laser excitation spectrum for Rb-Xe at T=453 K. For this experiment, $\lambda_2 = 794.7$ nm, [Xe] = $1.8 \times 10^{19}$ cm$^{-3}$, and $\Delta t = 8$ ns. Fourier transform of the spectrum is given in the right bottom corner.](image)
ence effect between the atomic or molecular states of the investigated gas mixture. This work, therefore, attributes these oscillations to quantum beating (Rabi oscillations).

It is worth mentioning that similar oscillations were observed on the red side of the K D₂ line when pumping the KXe gas mixture with circularly polarized light (Section 3.4.3).

3.3.4 Detection of Stimulated Raman Scattering by Two Color Excitation

Experiments show that two color pumping of various alkali noble gas mixtures in the vicinity of the Rb D₁ and D₂ lines induces stimulated Raman scattering (SRS). This effect may be observed indirectly by measuring the Rb D₂ line excitation spectra which are significantly suppressed when the difference between the two pumping wavelengths \( \Delta E = \lambda_2 - \lambda_1 \) equals the energy difference between the \(^2P_{3/2}\) and \(^2P_{1/2}\) states of alkali (\( \Delta E = 237 \text{ cm}^{-1} \) for Rb). The effect is described by an energy diagram given in Fig. 3.21. The diagram shows the stimulation of the Stokes components; however, it is also possible that the discussed suppression effect may be caused by the stimulation of the anti-Stokes component (red and blue arrows, pointing from and to a virtual state [dashed line], respectively, should be reversed).

Wavelengths \( \lambda_1 \) and \( \lambda_2 \) should be slightly detuned from the Rb D₂ and D₁ transitions in order to avoid strong absorption and radiation trapping processes. The pump wavelengths may be detuned by more than 2 and 3 nm from the line centers of the Rb D₁ and D₂ lines, respectively. Pump wavelengths may also be shifted to the blue side of the Rb D lines; however, a rapid decrease in the absorption coefficient of the blue side of the Rb D₂ line quickly suppresses the entire photoexcitation spectrum and makes it impossible to observe the SRS effect.

Excitation spectra for both Rb D₁ and D₂ are shown in Fig. 3.22 and 3.23, respectively. The ASE suppression peaks moves as a function of \( \lambda_1 \) and \( \lambda_2 \) always satisfying \( \frac{1}{\lambda_2} - \frac{1}{\lambda_1} = 237 \text{ cm}^{-1} \).

The demonstrated effect does not introduce new physics as stimulated Raman scattering is a well known phenomenon in alkali vapor [29]. However, this method may allow one to study the efficiency of the stimulated Raman scattering processes and, therefore, \( \chi^{(3)} \) as a function of \( \lambda_1 \) and \( \lambda_2 \) in the vicinity of the atomic reso-
Figure 3.21: Energy diagram demonstrating the detection of the stimulated Raman scattering via observation of the Rb D$_2$ line ASE suppression. Blue and red arrows represent the first ($\lambda_1$) and the second ($\lambda_2$) pump lasers, respectively.

Stimulated Raman scattering was observed in all tested alkali-noble gas mixtures (Rb-Xe, Rb-Ar and Rb-Kr). Moreover, the effect is not limited by alkali-noble gas systems and may be easily implemented using any other molecular system, provided that the two pump lasers are able to produce stimulated emission in a similar fashion.
Figure 3.22: Photoexcitation spectra demonstrating stimulated Raman scattering in RbXe gas mixture at $T=463$ K and $[\text{Xe}] = 1.8 \times 10^{19}$ cm$^{-3}$.

Figure 3.23: Photoexcitation spectrum demonstrating stimulated Raman scattering in RbXe gas mixture at $T=463$ K, $[\text{Xe}] = 1.8 \times 10^{19}$ cm$^{-3}$, and $\lambda_2 = 795.17$ nm.
3.4 Circularly Polarized Pump and Hyperfine 
Molecular and Atomic Structure

3.4.1 ASE Experiments

The original idea of using circularly polarized light to pump alkali-noble gas 
pairs arose because of the overlap of the Rb two photon absorption peak with 
the peak of the RbXe blue satellite (Fig. 3.4). As mentioned in Section 2.4, the 
$7^2S_{\frac{1}{2}} \leftarrow 5^2S_{\frac{1}{2}}$ two photon transition is forbidden in the case of circularly polar-
ized light. Therefore, pumping Rb-Xe diatomic pairs by circularly polarized light 
should yield excitation spectra which are not affected by the two photon absorp-
tion.

In this experiment, the setup introduced in Fig. 3.3 was used with minor 
changes. A high reflecting mirror was replaced with a beam block, the output 
coupler was removed, and a quarter wave plate was placed between the gas cell 
and a polarizing beam splitter. Figure 3.24 shows the excitation spectra taken 
at $T = 473$ K, pump energy of 1.3 mJ, and Xe pressure of 500 Torr. Red and 
black dots on the graph represent linear and circular polarizations of the pump 
laser, respectively. This experiment shows that circularly polarized light com-
pletely suppresses the two photon $7^2S_{\frac{1}{2}} \leftarrow 5^2S_{\frac{1}{2}}$ transition in the Rb vapor, as 
expected. Surprisingly, it also doubles the ASE efficiency without any change in 
the absorbed pump energy compared to the linearly polarized case.

The original experimental setup granted access to only one polarization com-
ponent of the alkali D$_2$ line ASE, which was not enough to understand the reason 
for the ASE efficiency dependence on the polarization of the pump. Additional 
experiments were required to explain this behavior.

An improved experimental setup was built in order to gain access to both po-
larization components of the ASE. The modified setup is shown in Fig. 3.25. The 
linearly polarized dye laser output (red dots represent vertically polarized light, 
normal to a plane of the experiment) was sent through a 50:50 non-polarizing 
beamsplitter and a quarter wave plate, and then entered the gas cell. Depend-
ing on the angle of the quarter wave plate, the pump polarization was kept un-
changed or transformed to circular polarization. The counter-propagating ASE 
pulse, emerging from the cell, passed through the same quarter wave plate, and 
was partially (50%) reflected by a non-polarizing beamsplitter. The reflected part
of the ASE was decomposed into the two orthogonal linear polarizations with a polarizing beamsplitter. Absolute energies of both polarizations were measured by two identical pyroelectric energy meters, designated as detector 2 and detector 3 for horizontal and vertical ASE polarization components, respectively.

The described setup was used to pump a RbXe gas cell filled with 600 Torr of Xe. A higher Xe pressure, compared to the previous experiment, was chosen in order to increase the ASE signal strength.

The gas cell was, at first, pumped with linearly polarized light (the main axis of the quarter wave plate was positioned parallel to the initial dye laser’s polarization). Both energy meters read similar values of the Rb D₂ line ASE energy (Fig. 3.26), indicating that the ASE had no preferable polarization. When the RbXe cell was pumped with circularly polarized light (the angle between the main axis of the quarter wave plate and the initial polarization of the dye laser was 45°) under the same conditions, the situation significantly changed (Fig. 3.27).

The horizontal polarization component (detector 2 in Fig. 3.25) of the ASE
increased by almost a factor of 2, and the vertical polarization component (detector 3) decreased by \( \sim 20\% \) compared to the case of linearly polarized pumping. This result may be explained under the assumption that some portion of the ASE, emerging from the gas cell, was circularly polarized. Furthermore, the direction of the polarization vector rotation was opposite to that of the pump beam because detector 2 measured the polarization component (reflected by the polarizing beam splitter) orthogonal to the polarization of the pump.

In order to confirm the assumption about the circularly polarized ASE, the quarter wave plate was placed before the NPBS. Such an arrangement enables pumping the gas cell with circularly polarized light without transforming the circularly polarized component of the ASE back to linear polarization. In this experiment both detectors 2 and 3 measured similar energies (as expected for circularly polarized ASE). Thus, data obtained in this and previous experiments demonstrates that when the RbXe gas mixture is pumped through the blue satellite utilizing circularly polarized light, some portion of the generated ASE is also circularly polarized.

It should be pointed out that the ratio of the energy values measured by detectors 2 and 3 (Fig. 3.27) does not represent the ratio of circularly to randomly polarized light. Detectors 2 and 3 measure equal amounts of randomly polarized ASE;
therefore, the relative portion of a circularly polarized component may be roughly calculated as:

\[ R_{\bigcirc} = \frac{E_2 - E_3}{E_2 + E_3}, \]  

(3.3)

where \( E_2 \) and \( E_3 \) are the energies measured by detector 2 and 3, respectively. Reflectance and transmittance of 50:50 non-polarizing beam splitters may significantly differ for s and p polarizations. This should be taken into account in Eq. 3.3 to obtain \( R_{\bigcirc} \) with better precision.

A similar set of experiments has been performed using the CsXe (blue satellite peaks at \( \sim 843.7 \) nm) gas mixture with the Xe pressure of 800 Torr. The photoexcitation spectra showing the measured values of two orthogonal polarization components of the Cs \( D_2 \) line (852.1 nm) ASE are given in Figs. 3.28 and 3.29 for the linearly and circularly polarized pump, respectively.

In the case of the CsXe gas mixture, the effect of the circularly polarized ASE
is significantly enhanced in contrast to the RbXe mixture. More than 70% of the emitted ASE was circularly polarized. The randomly polarized ASE component was suppressed by almost a factor of 4 (Fig. 3.29) compared to the data obtained while pumping with the linearly polarized pump (Fig. 3.28).

In order to characterize the effect of the circularly polarized pump in detail, efficiencies of the Rb and Cs D$_2$ line ASE pulses have also been measured as a function of the absorbed pump energy. The wavelength of the pump laser was tuned to the peak of the blue satellite of the corresponding alkali-noble gas molecule. Graphs showing the slope efficiencies of the Cs D$_2$ line ASE signals will be discussed further in this section due to the fact that the CsXe gas mixture exhibits a stark discrepancy between the circularly and randomly polarized ASE components in contrast to the RbXe mixture (efficiency of the RbXe gas mixture also was increased). Figure 3.30 shows the ASE efficiency plot as a function of the
absorbed energy. Considering that the NPBS had a reflectivity < 50% for p polarization, the underestimated value of the ASE efficiency turns out to be ~5% for only one side of the cell, which is considered to be a high value for a pulsed system.

As can be seen from Fig. 3.30, the lasing threshold values for the circularly polarized and non-polarized ASE components also differ from each other. This difference becomes more distinct if the efficiencies are plotted versus the energy of the pump laser (Fig. 3.31). The pump energy threshold values were measured to be ~ 470 µJ and 300 µJ (ratio of the lasing threshold values is ~3:2) for the non-polarized and circularly polarized ASE, respectively, implying that the population inversion between the hyperfine levels of the $6^2P_{3/2}$ and $6^2S_{1/2}$ states with $\Delta m_J=\pm 1$ is established faster than for other transitions.

As mentioned previously, the fact that detector 2 (polarization orthogonal to
the initial polarization of the pump light) measures greater energies compared to detector 3 suggests that the polarization vector rotation of the D$_2$ line ASE is opposite to that of the pump laser.

3.4.2 Circularly Polarized Alkali Lasers (CPALs)

Experimental results, discussed in the previous section, were used to design and demonstrate the first Circularly Polarized Alkali Laser (CPAL).

To the best of my knowledge, polarized laser emission from high temperature (T>450 K) gaseous gain media in the absence of external electric and magnetic fields (excluding the fields produced by the pump laser itself) has never been observed before. The reason is that a large number of collisions (∼ $10^9$ Torr$^{-1}$ s$^{-1}$ at the room temperature) occur in the gas phase and completely scramble the net
Figure 3.30: Dependence of ASE from the Cs D₂ line on energies absorbed by the pump. For all measurements Xe pressure was 800 Torr, T = 473 K, λ\textsubscript{pump} = 842.7 nm.

polarization of atoms and molecules before the radiative emission occurs (even if the system was initially pumped with a polarized light source).

Various optical elements (gratings, polarizers, polarizing beam splitters, windows tilted under the Brewster’s angle, etc.) are typically used to produce polarized light from gas lasers, but the gaseous gain medium does not have a preferred polarization direction. Therefore, the optical elements always selectively force the polarization direction and never result in an increase of the laser’s efficiency, but, typically, have the opposite effect (because of spectral hole burning and other effects).

The proposed, novel gas laser scheme allows one to generate the circularly polarized laser emission from the alkali D₁ and D₂ lines, with 50% increase in efficiency, and 34% decrease in the pump energy threshold compared to similar systems pumped with linearly polarized light. This is one of the most important re-
Figure 3.31: Dependence of ASE from the Cs D$_2$ line on the pump laser energies. For all measurements Xe pressure was 800 Torr, $T = 473$ K, $\lambda_{pump} = 842.7$ nm.

Results described in this dissertation because it demonstrates a new type of laser (selectively lasing on the transitions between certain hyperfine atomic states) which exhibits significantly better performance compared to its analogs with essentially no change in cost and pumping requirements.

The CPAL experimental arrangement is shown in Fig. 3.32. The initial pump laser had vertical polarization and the wavelength corresponded to the peak of the alkali-noble gas blue satellite. The pump laser beam passed through the PBS, transparent for the vertical polarization, and was transformed into the circularly polarized light with the quarter wave plate, and entered the quartz cell filled with alkali-noble gas mixture. The pump beam made a single pass through the cell. An unabsorbed portion of the pump beam was transformed by the second quarter wave plate back into the vertical polarization and was rejected from the laser cavity. The pump residual was measured by a pyroelectric detector, placed after the second PBS, which allowed us to calculate energy deposited into the gain.
medium.

When the population inversion was established for both alkali atomic D transitions, the gain medium started producing stimulated emission having circular polarization opposite to the polarization of the pump laser. Therefore, both quarter wave plates transformed the circularly polarized laser emission into horizontally polarized light which was reflected by the PBSs into the high reflecting and output coupling mirrors, constituting a cavity, shown on the right and left sides of Fig 3.32, respectively.

The comparison of the efficiencies of the studied systems pumped with the circularly and linearly polarized laser is shown in Fig. 3.33 and 3.34 as a function of the absorbed pump energy and the total pump energy, respectively. In order to measure the efficiency of the linearly polarized pump, both quarter wave plates were removed from the laser cavity.

Efficiency plots given in Fig. 3.33 and 3.34 incorporate the output energies of both D1 and D2 lasers operating simultaneously. The D2 line laser efficiency was measured separately using a D2 line bandpass filter and was exactly 2/3 of the D1 + D2 efficiency. Thus, the Cs D2 line laser had an (emitted/absorbed) efficiency of 12.4% which is a high value considering the pulsed nature of the laser system and the fact that the cavity was not optimized (OC reflectivity is not optimal, many optical elements with no AR coating).

While the exact nature of the observed ASE efficiency enhancement when pumping with circularly polarized light is not thoroughly studied, the experimental data presented in this section is compelling. The results clearly demonstrate a significant enhancement in the alkali laser operation when pumped with circularly polarized light. The system described here is the most efficient (12.4%) atomic al-

![Figure 3.32: Experimental arrangement used for demonstration of circularly pumped alkali laser.](image-url)
Figure 3.33: Dependence of laser emission (system simultaneously lasing on D\textsubscript{1} and D\textsubscript{2} transitions) of Cs on energies absorbed by the pump. For all measurements Xe pressure was 800 Torr, T = 473 K, $\lambda_{\text{pump}} = 842.7$ nm. Black and blue dots represent results for the circularly and linearly polarized pump, respectively.

In summary, although the proposed arrangement looks similar to the original XPAL and DPAL setups (the only difference is a presence of two quarter wave plates between the gas cell and PBSs), it introduces several new physical effects, significantly changing the dynamics of this laser system and improving its characteristics with respect to lasing energy threshold and efficiency, without affecting overall pump energy, absorbed pump energy, and temperature of the gas mixture. It is strongly recommended to use the suggested pumping scheme in high power DPAL laser systems (pumped with CW or quasi-CW lasers). It is expected that the circularly polarized pump will also improve the slope efficiency and lasing threshold of DPALs, significantly increasing the impact of these systems on the
Figure 3.34: Dependence of laser emission (system simultaneously lasing on D\textsubscript{1} and D\textsubscript{2} transitions) of Cs on the pump laser energies. For all measurements Xe pressure was 800 Torr, T = 473 K, \( \lambda_{\text{pump}} = 842.7 \) nm. Black and blue dots represent results for the circularly and linearly polarized pump, respectively.

3.4.3 Observation of Quantum Oscillations on the D\textsubscript{2} Line Red Satellite of K-Noble Gas Pairs

Previous experiments show that a circularly polarized light induces a non-equilibrium population distribution between adjacent spin states. Recall, that previously observed in RbXe (Section 3.3.3) oscillations in proximity of the Rb D\textsubscript{2} line were classified as potential oscillation between atomic or molecular spin states.

It has been suggested to pump several alkali-noble gas mixtures with circularly polarized light and monitor the stimulated emission emerging from the gas cell by means of the setup shown in Fig. 3.25.

In this experiment KAr and KXe gas mixtures having noble gas pressures of
Figure 3.35: Laser excitation spectra for KXe measured using the setup given in Fig. 3.25 at Xe pressure of 300 Torr, $T = 495$ K, and pump energy of 3 mJ. Top and bottom figures show the values measured by detector 3 and detector 2, respectively. Fourier transform of the bottom spectrum is given in the right bottom corner.

300 Torr were pumped with circularly and linearly polarized light in the vicinity of the K D$_2$ line (766-768 nm). A relatively strong stimulated emission ($\sim 2 \mu J$) was generated in the direction opposite to the pump beam in both cases.

When the gas mixtures were pumped with circularly polarized light an oscillatory behavior similar to those observed in the two color pumping experiment was detected for the horizontal polarization component (detector 2) as shown in Fig. 3.35. Recall that difference in signals measured by energy detectors 2 and 3 (Fig. 3.25) suggests that the detected emission is partially circularly polarized. Therefore, only circularly polarized emission from the KAr and KXe gas mixtures exhibited oscillatory behavior. Moreover, no oscillations were detected in the case of linearly polarized pump light.

The frequency of the oscillations was $\sim 35.6$ GHz, which is similar to that
observed for RbXe (~ 30.9 GHz). The oscillations of the same frequency were measured from both KXe and KAr gas mixtures. It has been observed that the amplitude of the oscillations was greater in the case of KXe compared to other alkali-noble gases.

The described oscillations are probably similar in nature to those studied in [30]. This work does not investigate the observed effect thoroughly but only demonstrates its presence and suggests that future studies should be conducted in order to identify the origin of these oscillations.
Chapter 4

TIME-DEPENDENT RATE EQUATION MODEL

The main goal of this simulation is to investigate the 3 to 2 laser level collapse effect observed in RbXe gas mixture in order to explain the physics behind it. This model ignores several physical processes which may cause some disagreement with theoretical data. For example, it is well known that the atomic alkali lasers pumped by the photoassociation of atomic pairs lase on both D$_1$ and D$_2$ lines of the alkalis; therefore, it is desirable to simulate both lasing transitions at the same time in order to obtain more reliable results. This simulation takes into consideration only the D$_2$ line laser. Nevertheless, the model provides a good understanding of the 3 to 2 laser level collapse effect that has never been theoretically described before and, therefore, fulfills its purpose. Several useful suggestions concerning future simulations and experiments are given at the end of this chapter. Basic modeling concepts of the pulsed laser’s behavior using rate equations were adapted from [31, 32, 33].

It is worth mentioning that the time-dependent rate equation modeling was done for the Cs-Ar and Cs-Ar-C$_2$H$_6$ XPALs by J. D. Readle in [34] using a different approach from the one described here. A combination of both models may significantly benefit future simulations.

4.1 Rate Equations Model For a Four-Level Laser System

The real laser system was modeled by taking into account its geometry, temperature, pump waveform profile and energy, resonator losses, and the mode volume of the optical cavity.

The simulated system is schematically shown in Fig. 4.1. States $|1\rangle$, $|2\rangle$, $|3\rangle$, and $|4\rangle$ represent the $5^2S_{\frac{1}{2}}$ (considering it to be the same as $X^2\Sigma^+_\frac{1}{2}$), $5^2P_{\frac{1}{2}}$, $B^2\Sigma^+_\frac{1}{2}$, and $5^2P_{\frac{3}{2}}$ states of RbXe molecule and Rb atom (Fig. 2.2), respectively. The
$5^2P_{\frac{1}{2}}$ state ($|4\rangle$ in Fig. 4.1) plays an important role in the process of the $5^2P_{\frac{1}{2}}$ ($|2\rangle$) depletion. The model does not take into account lasing on $|4\rangle \rightarrow |1\rangle$ transition but takes care of only spontaneous emission and relaxation processes.

First, a treatment of the optical cavity (Fig. 3.3), comprising an output coupling (OC) mirror, high reflecting (HR) mirror, polarizing beamsplitter (PBS), and a RbXe cell (gain medium), is described.

The refractive indexes of the gain medium and ambient air were approximated to be 1. As a result, an optical length of the resonator is equal to its physical length ($L$). The same is valid for the value of the optical length of the gain medium ($l$).

Assuming that the cavity mirrors have no internal losses (1 = $R + T$, where $R$ and $T$ are reflectance and transmittance, respectively), the intra-cavity laser intensity ($I'$) after one round trip of the cavity may be expressed in the following way:

$$I' = I R_1 R_2 (1 - T_i)^2 \exp(2 \sigma \Delta N l),$$

where $R_1$ and $R_2$ are the reflectivities of the OC and HR mirrors, respectively, $1 - T_i$ is the single pass internal loss of the cavity, $\sigma$ is a stimulated emission cross-section (corresponding to the laser transition $|2\rangle \rightarrow |1\rangle$), and $\Delta N$ is the population inversion ($\Delta N = (N_2 - N_1 \frac{g_2}{g_1})$, where $N_1, N_2$ and $g_1, g_2$ are the populations and degeneracy factors of state $|1\rangle$ and $|2\rangle$, respectively).

It is convenient to express losses associated with OC, HR, and Fresnel reflections from the cell’s windows and PBS in a logarithmic notation as $\gamma_1, \gamma_2$, and $\gamma_i$. 

Figure 4.1: Energy level diagram of the modeled laser system.
respectively:

\[ \gamma_1 = -\ln(R_1), \quad (4.2) \]
\[ \gamma_2 = -\ln(R_2), \quad (4.3) \]
\[ \gamma_i = -\ln(1 - T_i). \quad (4.4) \]

They may be combined in the total logarithmic loss per pass:

\[ \gamma \equiv \gamma_i + \frac{\gamma_1 + \gamma_2}{2}. \quad (4.5) \]

Using the introduced logarithmic notation, one of the most important characteristic values of the cavity, the photon lifetime \((\tau_c)\), may be calculated as:

\[ \frac{1}{\tau_c} = \frac{\gamma c}{L} + \frac{\gamma_1 c}{2L} + \frac{\gamma_2 c}{2L} = \frac{\gamma c}{L}, \quad (4.6) \]

where \(c\) is the speed of light.

Another important number is a volume of the mode inside the gain medium \((V_a)\). Knowing that the cavity is longitudinally pumped with a collimated, uniform laser beam of diameter \(d\), the expression for the modal volume may be approximated as:

\[ V_a = \frac{\pi d^2}{4} l. \quad (4.7) \]

At this point, we are going to return to the calculation of the laser’s intensity in the cavity. With the use of the logarithmic notation for losses and slightly rearranging Eq. 4.1 we arrive at:

\[ \Delta I = (\exp[2(\sigma \Delta N l - \gamma)] - 1) \, I, \quad (4.8) \]

where \(\Delta I \equiv (I' - I)\) is the difference between initial laser intensity and its intensity after one cavity round trip. Assuming that:

\[ \sigma \Delta N l - \gamma \ll 1, \quad (4.9) \]

and expanding the exponential as a power series, Eq. 4.8 becomes:

\[ \Delta I = 2(\sigma \Delta N l - \gamma) I. \quad (4.10) \]

Dividing Eq. 4.10 by the round trip time, \(\Delta t = 2L/c\), and assuming that
\[ \Delta I / \Delta t \simeq dI / dt: \]
\[ \frac{dI}{dt} = \left( \frac{\sigma l c}{L} \Delta N - \frac{\gamma c}{L} \right) I. \]  

(4.11)

To simplify Eq. 4.11 we define a stimulated transition rate coefficient \( B \) and the mode volume in the cavity \( V \):
\[ B = \frac{\sigma l c}{L V_a}, \]  

(4.12)
\[ V = \frac{L V_a}{l}. \]  

(4.13)

It can also be recognized that the second term in Eq. 4.11 is the inverse of the cavity lifetime from Eq. 4.6. Finally, combining Eqs. 4.6, 4.11 - 4.13 and recalling that intensity is proportional to the number of photons \( \phi \), an expression showing the evolution of the photon number in the cavity as a function of time may be written as:
\[ \frac{d\phi}{dt} = \left( B V_a \Delta N - \frac{1}{\tau_c} \right) \phi. \]  

(4.14)

The stimulated emission cross-section \( \sigma \) is related to Einstein’s coefficient \( A_{21} \), wavelength corresponding to the current transition \( \lambda_{21} \), and the value of the line shape function \( g(\nu_{21}) \) at \( \nu_{21} = c/\lambda_{21} \) via the following famous expression:
\[ \sigma(\nu) = A_{21} \frac{\lambda_{21}^2}{8\pi} g(\nu). \]  

(4.15)

The line shape function has a peak value:
\[ g(\nu_{21}) = \frac{2}{\pi \Gamma}, \]  

(4.16)
assuming a Lorentzian distribution having a width \( \Gamma \).

Finally, the output power of the laser is calculated as:
\[ P_{out} = \frac{\gamma_{21} c}{2L} \hbar \nu_{21} \phi, \]  

(4.17)
where \( \hbar \nu_{21} \) is the energy of the photon corresponding to the laser \( \langle 2 \rightarrow 1 \rangle \) transition

A system of rate equations for each level shown in Fig. 4.1 can now be written by using the quantities defined earlier in this section. The system comprises five differential equations. Four of them describe all transitions given in Fig. 4.1 and
the last one defines the evolution of the number of photons in the optical cavity as a function of time (Eq. 4.14):

\[
\begin{align*}
\frac{dN_1}{dt} &= -\sigma_{abs}R_{pump}(N_1 - N_3) + \frac{N_2}{\tau_{21}} + V_a B\phi(N_2 - N_1 \frac{g_2}{g_1}) + \frac{N_4}{\tau_{41}}, \\
\frac{dN_2}{dt} &= -\frac{N_2}{\tau_{21}} - V_a B\phi(N_2 - N_1 \frac{g_2}{g_1}) + \frac{N_3}{\tau_{32}} - \frac{N_2}{\tau_{24}}, \\
\frac{dN_3}{dt} &= +\sigma_{abs}R_{pump}(N_1 - N_3) - \frac{N_3}{\tau_{32}}, \\
\frac{dN_4}{dt} &= +\frac{N_2}{\tau_{24}} - \frac{N_4}{\tau_{41}}, \\
\frac{d\phi}{dt} &= +V_a B(\phi + 1)(N_2 - N_1 \frac{g_2}{g_1}) - \frac{\phi}{\tau_c}.
\end{align*}
\]

(4.18)

In this system, \(\sigma_{abs}\) is the absorption cross-section for the \(|3\rangle \leftarrow |1\rangle\) transition which corresponds to the peak of the RbXe D\(_2\) line blue satellite (\(B^2\Sigma_1^+ \leftarrow X^2\Sigma_2^+\)), and \(R_{pump}\) is the number of pumping photons per unit area.

The temporal profile of the pump pulse was approximated by the Gaussian distribution function with \(\sigma_p = 3.8 \times 10^{-9}\) peaking at \(t_0 = 20\) ns:

\[
P(t) = \frac{1}{\sigma_p \sqrt{2\pi}} e^{-\frac{(t-t_0)^2}{2\sigma_p^2}}.
\]

(4.19)

There are several comments to be made on the system of equations 4.18. First, in the last (5th) equation one extra photon has to be added to the total number of photons in the cavity. This takes care of the problem associated with the absence of initial photons in the cavity when the population inversion reaches its threshold. Note that \(d\phi/dt\) can be negative; however, if we interpret \(\phi\) as being the number of photons in the cavity then it must be a non-negative quantity. Therefore, the situation when \(\phi < 0\) which often emerges in the simulation is discarded to avoid misleading results.

The degeneracies of state \(|1\rangle\) and \(|3\rangle\) are equal and, therefore, the absorption rate is proportional to \(N_1 - N_3\). At the same time, degeneracy of state \(|2\rangle\) is twice that of \(|1\rangle\). As a result, the emission rate is proportional to \(N_2 - 2N_1\).
4.2 Evaluation of Model: Comparing Simulated and Experimental Results

The accuracy of the described model was tested by comparing the temporal profiles, lasing threshold energies, and efficiencies of the simulated and experimentally studied laser systems. The temperature dependencies of the alkali vapor pressure were taken from [23] (Fig. 3.2). The physical quantities used in this simulation are given in Table 4.1. Two adjusting parameters were the lifetimes $\tau_{24}$ and $\tau_{41}$ which may slightly deviate from the real values due to the fact that lasing on $|4\rangle \rightarrow |1\rangle$ is not taken into account by this model.

The simulation shows a good agreement between the calculated Rb D$_2$ line laser pulse shape (Fig. 4.2 bottom graph) and the waveforms experimentally measured (Fig. 4.2 upper graph) using high-speed silicon photodetectors (rise time $< 1$ ns). The calculated laser efficiency was also in agreement with the experimental data (Fig. 4.3) for $T < 440$ K (the value of $\frac{\Delta E}{kT}$ > 1.1). The laser efficiency plot demonstrates that the current rate equation model is able to calculate the absolute values of the output energy of the laser with high precision. For higher temperatures the simulated results start deviating from the experimentally obtained data. These deviations emerged due to the collisional coupling (between states $|2\rangle$ and $|3\rangle$) which becomes non-negligible when $\frac{\Delta E}{kT} < 1.1$ (beginning of the collapse of the 3 level into 2 level system), meaning that additional equations for adequately

<table>
<thead>
<tr>
<th>Table 4.1: Constants used in the rate equation model</th>
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<tbody>
<tr>
<td><strong>Value</strong></td>
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<tr>
<td>--------------------------</td>
</tr>
<tr>
<td>Pump wavelength ($\lambda_{13}$)</td>
</tr>
<tr>
<td>Laser wavelength ($\lambda_{21}$)</td>
</tr>
<tr>
<td>Absorption cross-section ($\sigma_{21}$)</td>
</tr>
<tr>
<td>HR mirror reflectance, ($R_1$)</td>
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<tr>
<td>OC mirror reflectance, ($R_2$)</td>
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<tr>
<td>Single pass internal transmission of the cavity, ($T_i$)</td>
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<td>Resonator length, ($L$)</td>
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<td>Gain medium length, ($l$)</td>
</tr>
<tr>
<td>Pump beam diameter, ($d$)</td>
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<tr>
<td>Line width of state $</td>
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<td>Lifetime of state $</td>
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4.3 Modeling the Collapse of a Three Level Laser into a Two Level System

The system collapse is associated with collisional coupling of the upper laser state ($|2\rangle$) and the upper pump state ($|3\rangle$). The first, simplest option to simulate this coupling is to assume that the population of state $|3\rangle$ not only depends on the
transition rate \( R_{32} \):
\[
R_{32} = \frac{N_3}{\tau_{32}},
\]  
but also on the excitation transition rate \( R_{23} \) of the system from state \( |2\rangle \) to state \( |3\rangle \), which is dependent on \( \frac{\Delta E}{kT} \). Assuming that by analogy with \( R_{32} \) this transition rate is proportional to the population of state \( |2\rangle \), it may be written as:
\[
R_{23} = \frac{N_2}{\tau_{32}} \eta \left( \frac{\Delta E}{kT} \right),
\]  
where \( \eta \left( \frac{\Delta E}{kT} \right) \) is a coefficient responsible for the growth of the transition rate \( R_{23} \) with increase of the temperature. In other words, Eq. 4.21 includes terms for the growth of the upper state population in terms of its excited state lifetime (\( \tau_{32} \)) and system temperature.

The energy difference (\( \Delta E \)) between \( |3\rangle \) and \( |2\rangle \) is constant if the pump wavelength is fixed, and the collisional line broadening (which is much smaller than the
value of $\Delta E$) is assumed to be constant with temperature. Therefore, $\eta$ becomes a function of temperature only and, in the future, will be designated as $\eta(T)$.

Introduction of $R_{23}$ to the current rate equation model is effectively equivalent to raising of the upper state, $|3\rangle$, lifetime. This unavoidably leads to a slight increase of the pump energy threshold value (as $\eta(T)$ increases) which was not observed in the experiment (Fig. 3.5). This occurs due to the dependence of the lifetimes (Table 4.1) on the temperature which was not taken into account by this model. Nevertheless, the suggested model is still able to provide a qualitative description of physical processes occurring in the system when it approaches the critical point ($\Delta E/kT \leq 1.1$).

Figure 4.4 shows the calculated $\eta$ coefficients required to match the experimental data on the RbXe gas mixture. An overlap of these coefficients for the different values of the pump energies would indicate that the model may ideally describe the system collapse process. The coefficients, however, slightly differ from each other, which will cause some mismatch in the experimental and calculated values of the laser output energies.

The first model assumes that the transition rates $R_{32}$ and $R_{23}$ depend only on the population of $|3\rangle$ and $|2\rangle$ respectively. It is also possible that the actual population balance between states $|3\rangle$ and $|2\rangle$ is driven by a coefficient proportional to the ratio of their populations $\frac{N_3}{N_2}$. Taking this into consideration, the second model is based in the assumption that when the ratio $\frac{N_3}{N_2}$ reaches a certain value $\zeta(T)$ ($\zeta$ plays a role similar to that of $\eta$ in Eq. 4.21), the lifetime of state $|3\rangle$ becomes large enough to effectively nullify the transition rate $R_{32}$. The reduction of $R_{32}$ should, therefore, be described by a function $f\left(\frac{1}{\zeta(T)}\frac{N_3}{N_2}\right)$. It is natural to assume that this function has an exponential behavior, leading to a modified transition rate:

$$R_{32} = \frac{N_3}{\tau_{32}} \exp\left(\frac{1}{\zeta(T)}\frac{N_3}{N_2}\right). \quad (4.22)$$

The calculated $\zeta$ coefficients for the system described by the transition rate given in Eq. 4.22 are plotted in Fig. 4.5. Due to the fact that the $\eta$ coefficient behaves similarly to $\zeta$, and has a simpler functional form, $\eta$ (and Eq. 4.21)) was chosen for use in the current model.

The $\eta$ coefficient calculated for the pump energy of 7.7 mJ (red circles in Fig. 4.4) may be approximated with simple Maxwell-Boltzmann statistics in the following way:
\[ \eta(T) = \frac{\exp\left(\frac{-\Delta E}{kT}\right)}{0.21} - 1.57. \]  

(4.23)

Recall that \( \eta(T) \) in Eq. 4.21 is just a factor which is responsible for the change of the population transfer rate from state \(|2\rangle\) to state \(|3\rangle\) as a function of temperate. It should be proportional to the Maxwell-Boltzmann factor; however, it would be an extraordinary occurrence if this rate (which, in general, should depend on gas pressure, collisional cross-section and many other factors) were directly proportional to \( \exp\left(\frac{-\Delta E}{kT}\right) \). Thus, the scaling factors (0.21 and 1.57) in Eq. 4.23 should not be surprising. Furthermore, the fact that no additional functions are required to connect \( \eta(T) \) with the Maxwell-Boltzmann factor imply that the chosen way of describing the energy level collapse process is close to the reality, and, by improving the existent model, it is possible to achieve a better match between the theory and experiment.

Figure 4.4: Dependence of coefficients \( \eta \) as a function of the temperature, blue triangles, red circles and black squares represent pump energies of 10 mJ, 7.7 mJ, and 5.8 mJ, respectively.
Using the additional transfer rate component, $R_{23}$ (Eq. 4.21), with the $\eta(T)$ coefficient, described with Eq. 4.23, in the initial system of rate equations (Eq. 4.18) originates a result, given in Fig. 4.6.

As mentioned before, the calculated $\eta(T)$ coefficient varies slightly for different pump energies, which indicates that the proposed model forces the lasing threshold to increase with increasing temperature. In the end, the proposed time-dependent rate equation model predicts that the breakpoint (where the laser efficiency trend changes) is temperature-dependent (Fig. 4.6), which was not observed in the experiment. The simulation results, however, are not unreasonable in the sense that the higher pump energies should allow the laser system to operate at higher temperatures. In other words, infinite pump energy should allow a two level system to lase. Thus, no temperature dependence was observed in the experiment, most likely due to the fact that the lifetime of state $|2\rangle$ was also temperature dependent and compensated for the shift of the breakpoint.
Figure 4.6: Simulated (lines) and experimentally measured (circles) RbXe laser output as a function of the temperature. Blue, red, and black color represent pump energies of 10 mJ, 7.7 mJ, and 5.8 mJ, respectively.

It is, therefore, recommended to repeat the same experiment for several alkalis and different rare gas pressures. It is expected that the lasing threshold will be, in general, temperature dependent which will indicate that the current simulation results are adequate. The experimental data measured for various noble gas pressures will allow one to estimate the lifetime dependencies on the temperature and number of collisions per second, which may be taken into account in the current rate equation model. It is also desirable to simulate both D$_1$ and D$_2$ lasing transitions in the future to increase the precision of the model. Therefore, the energies of both these lasers should be measured independently in the future experiments.

In summary, this section describes the first simulation of the collapse of a 3 level laser into a 2 level system. The proposed time-dependent rate equation model demonstrates that the Maxwell-Boltzmann distribution is suitable for simulating the 3 to 2 level system collapse. The model gives a good agreement with experimental data; however, a more detailed modeling and additional experiments are required in order to provide better agreement between this simulation and experimental data.
The set of experiments described in this dissertation investigated multiple mechanisms enabling the study of fundamental physics of alkali-rare gas mixtures and also demonstrated novel pumping techniques of alkali-rare gas atomic pairs.

It has been demonstrated that the minimum energy separation, $\Delta E$, between two upper laser states in RbXe atomic laser systems pumped by the photodissociation of atomic pairs is $\sim 1 \text{ kT}$. This result is in good agreement with the value of $\Delta E \approx 0.7 \text{ kT}$ measured by means of a CsAr system using a different technique [9]. This dissertation also described the first simulation of the laser system collapse utilizing a time dependent rate equation model.

Further, this work introduced and studied two excitation methods of the alkali vapor-noble gas mixtures: two color pumping and pumping with circularly polarized light. Both of these methods were used to perform a variety of experiments, some of which were described in Chapter 3.

The two color pumping experiment performed using RbXe gas mixture has revealed the presence of a small ($\sim 6 \text{ cm}^{-1}$) energy barrier on the $A^2\Pi_1^+$ molecular state that has never been observed experimentally. Pumping the Rb atoms through this barrier allowed lasing on Rb D$_2$ transition with (optical-to-optical) quantum efficiency above unity, converting some portion of the kinetic energy of the atoms into the energy of the emitted photons. It has been demonstrated that the same energy barrier may be implemented for amplification of light utilizing free $\rightarrow$ free molecular transitions. It has also been shown that two color pumping allows the investigation of stimulated Raman scattering and quantum beating effects in alkali vapor-noble gas mixtures. These effects were observed, but further investigation is required for more thorough characterization.

Another excitation method, pumping with circularly polarized light, has been used for decades to produce hyperpolarized noble gas atoms. This work, however, demonstrated that circularly polarized light (exciting molecular transitions) may be used to induce stimulated emission (also circularly polarized) on alkali atomic
transitions between specific hyperfine states (in particular, states with $\Delta m = \pm 1$). Such an excitation process improves the efficiency of the CsXe and RbXe atomic laser systems by 50% due to the fact that only one spin state in the $5^2P$ and $6^2P$ manifold of Rb and Cs atoms, respectively, experiences significant population inversion with respect to ground. It has also been demonstrated that circularly polarized light induces quantum beating similar to that observed with two color pumping. Excitation of molecular alkali-noble gas transitions, in contrast to atomic transitions which are currently used, may also benefit the production efficiency of hyperpolarized noble gas atoms used in science and medicine.

It is expected that the methods and experiments described in this dissertation will be further developed and implemented in systematic studies of fundamental physics and robust atomic and molecular laser systems.
References


68


