Abstract

The research project to be performed by the United States Air Force Academy SPS chapter aims at measuring collisional excitation transfer and quenching cross-sections in rubidium-methane and rubidium-deuterated methane systems. This study is relevant for alkali laser development and for understanding collisional processes in alkali-buffer gas systems.
Overview of Proposed Project

The proposed research project aims at studying collisional processes in rubidium-buffer gas systems subjected to laser radiation. When a rubidium atom is excited from the ground state to the \(5P_{3/2}\) state by a 780 nm photon in the presence of a buffer gas, several processes can occur: (1) spontaneous (radiative) decay to the ground state and emission of a 780 nm photon, (2) collisional quenching (non-radiative decay), and (3) collisional excitation transfer to the \(5P_{1/2}\) state followed by emission of a 795 nm photon.

The third process is at the heart of the operation of an alkali laser, a laser whose active medium is an alkali vapor such as rubidium, cesium or potassium. The first two processes act as losses in the laser gain medium. While the radiative decay is a natural process with rates that are well known, the other two processes depend on the alkali-buffer gas combination and are not well understood.

The goal of the proposed research project is to measure collisional excitation transfer and quenching cross-sections in rubidium-methane and rubidium-deuterated methane systems. This study is relevant for alkali laser development and for understanding collisional processes in alkali-buffer gas systems.

The proposed project will provide hands-on experience to three undergraduate students. This is particularly important at an undergraduate institution such as USAFA, as our cadets become officers in the US Air Force immediately upon graduation and are often posted to technical positions where a scientific background is extremely useful. To provide undergraduates with meaningful research experience is one of the SPS National objectives that our SPS chapter fully embraces.

Background for Proposed Project

An alkali laser has the capability of delivering extremely high powers with excellent beam quality making this type of laser desirable in a variety of applications [1]. Methane (\(\text{CH}_4\)) is highly pursued as a buffer gas, due to its favorable collisional excitation transfer cross-section. However, recent experiments aimed at scaling the output laser power to higher levels showed a degradation of the laser performance over time, caused by a decrease in laser efficiency and eventually termination of laser action [2]. Further studies attributed this power degradation to thermal effects in the laser gain medium due to collisional quenching [3]. Other studies hinted at deuterated methane (\(\text{CD}_4\)) being a better candidate than \(\text{CH}_4\) due to its lower collisional quenching cross-section [4]. Large uncertainties associated with excitation transfer and quenching cross-sections on the order of 15\% and 30-50\%, respectively, were reported [4]. It is also interesting to note that some estimates of the collisional excitation transfer cross-section are inferred from the performance of the alkali laser under various experimental conditions (buffer gas pressure and temperature) and not directly measured [5].

The proposed project entails measuring these cross-sections with high precision. We have previously measured the excitation transfer cross-section in Rb-He with a 2\% uncertainty [6]. This is another alkali-buffer gas combination pursued, since quenching is essentially negligible, even though the excitation transfer cross-section is not as high. We reported the first time observation of a quadratic dependence of the collisional excitation transfer rate on the buffer gas pressure (previously believed to be linear) and we interpreted this enhancement as being due to three-body collisions between one Rb and two He atoms [7]. We also reported the first experimental study of the temperature variation of the three-body transfer rate and developed a theoretical model to explain our experimental results [8]. To further test our model and better understand the three-body process we are currently investigating buffer gas mixtures such as He-Ar and He-Xe.
The goal of the proposed research project is to determine collisional excitation transfer and quenching cross-sections in Rb-CH\textsubscript{4} and Rb-DH\textsubscript{4} systems. As described in detail below, this will be accomplished by measuring the collisional excitation transfer and quenching rates as a function of the buffer gas pressure and fitting the data to known functional forms to extract the desired cross-sections.

### Description of Proposed Research - Methods, Design, and Procedures

In the experiment, a self-mode-locking Ti:Sapphire laser creates ultrafast laser pulses at 780 nm which excite Rb atoms to the 5P\textsubscript{3/2} state. These Rb atoms collide with a buffer gas, either CH\textsubscript{4} or CD\textsubscript{4}, at a given pressure. The buffer gas facilitates a transition from the 5P\textsubscript{3/2} state to the 5P\textsubscript{1/2} state and the resulting photons (795 nm) are detected by a photomultiplier tube. These fluorescence photons are recorded as a function of time by a time-to-digital converter.

The time evolution of the 5P\textsubscript{3/2} state population after termination of the laser pulse is described by the following rate equation:

$$\frac{dn_2}{dt} = -(\gamma_2 + R_{21} + Q_2)n_2 + R_{12}n_1$$

where $n_2$ is the population of the 5P\textsubscript{3/2} state, $n_1$ is the population of the 5P\textsubscript{1/2} state, $\gamma_2$ is the spontaneous decay rate of the 5P\textsubscript{3/2} state, $R_{21}$ is the collisional transfer rate from the 5P\textsubscript{3/2} state to the 5P\textsubscript{1/2} state, $R_{12}$ is the collisional transfer rate from the 5P\textsubscript{1/2} state to the 5P\textsubscript{3/2} state, and $Q_2$ is the collisional quenching rate of the 5P\textsubscript{3/2} state. The 5P\textsubscript{1/2} state population is modeled by a similar rate equation. Solving the coupled differential equations gives a solution in the form of a double exponential:

$$n_1(t) = Ae^{s_+t} + Be^{s_-t}$$

where $A$, $B$, $s_+$, and $s_-$ are coefficients depending on $\gamma$, $R$, and $Q$. By fitting the data we collected to this functional form and knowing $\gamma$, we can then determine $R$ and $Q$ at that pressure. An example data set obtained for 500 Torr of He is shown in Fig. 1.

The rising portion of the curve is due to collisional excitation transfer, while the decay portion is due to spontaneous decay. For inert gases such as He, quenching is essentially negligible, but becomes important in molecular systems such as CH\textsubscript{4} or CD\textsubscript{4}, due to extra degrees of freedom. Quenching in this case results in a faster decay to the ground state.

The experiment will be repeated for various buffer gas pressures up to 100 Torr. The collisional excitation transfer $R$ and quenching $Q$ rates thus determined will be plotted as a function of the buffer gas pressure. The slope of each curve will determine the respective cross-section.

![Fig. 1. Rb fluorescence due to collisional excitation transfer in 500 Torr of He](image-url)
Plan for Carrying Out Proposed Project

- **Personnel** – Cadet First Class Jeremiah Wells will be the leader of the proposed research project. He is a senior physics major with a concentration in “Lasers and Optics” and an SPS member. Two junior physics majors will be involved in this project. They will work under the supervision of C1C Wells during the spring semester and they will ensure the continuity of the project once C1C Wells graduates in May 2017. One of the two juniors is currently an SPS member and it is expected that the other one will join SPS National as well.

- **Expertise** – C1C Wells is currently working on a research project involving measurements of collisional fine-structure excitation transfer cross-sections in Rb-inert buffer gas mixtures such as He-Ar and He-Xe. The proposed project will use the same experimental setup that C1C Wells is familiar with. He has experience with the operation of the mode-locked laser, the vacuum, photon detection and LabView-based data acquisition systems, as well as the data analysis.

- **Research space** – The research project will be carried out in the Laser and Optics Research Center (LORC) at the US Air Force Academy. The experimental setup is already in place in one of the research laboratories designated for this purpose.

- **Contributions of faculty advisors or the department** – Dr. Alina Gearba, the SPS faculty advisor, and Dr. Jerry Sell, a research scientist in the LORC, will serve as project mentors. They have extensive experience with lasers and optics and published three papers on this topic. The LORC will provide the laboratory space and the experimental setup.

### Project Timeline

There will be a learning curve for the new project members to become familiar with the apparatus and the technique, but nevertheless, we expect to adhere to the following timeline:

- **Mid-May 2017** – methane study completed
- **May 31, 2017** – interim report submitted
- **June 5-9, 2017** – results disseminated at the Annual Meeting of the Division of Atomic, Molecular and Optical Physics of the APS in Sacramento, CA
- **Mid-August 2017** – manuscript submitted for publication describing the methane study
- **October 27-28, 2017** - results disseminated at the Annual Meeting of the Four Corners of the APS in Fort Collins, CO
- **Mid-December 2017** - deuterated methane study completed
- **December 31, 2017** - final report submitted
- **Mid-March 2018** - manuscript submitted for publication describing the deuterated methane study
Budget Justification

The funding we requested will be used to purchase pressurized bottles of CH₄ and CD₄ at $305 and $1230, respectively, to perform the proposed experiments. This study will answer the question as to whether CD₄ is a better candidate than CH₄ as buffer gas for an alkali laser as previously speculated. Therefore, the requested funding is essential to the successful completion of the proposed study.

The proposed research project will be carried out in the Laser and Optics Research Center (LORC) at the US Air Force Academy in one of the research laboratories designated for this purpose. The experimental setup is already in place and includes the self-mode-locking Ti:Sapphire laser, vacuum system, photomultiplier tube, time-to-digital converter, and associated optics and electronics.

Bibliography