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Laser-slowing and trapping of molecules for ultracold chemistry

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14. ABSTRACT
In this project, we will investigate collisions between cold homonuclear molecules held in a magneto-optical trap, both theoretically and experimentally. The experiment will be carried out at Prof. Marcassa's group, at University of São Paulo in São Carlos. The main goal in this project is to study collisions in a cold trapped molecular sample in a well-defined quantum state. Such collisions will involve atom-molecule and molecule-molecule collisions in homonuclear samples (Rb?Rb2 and Rb2-Rb2). Dr. Bouloufa's group will carry out molecular
structure calculations which will be implemented into various numerical codes for dynamical calculations (grid methods, wave packet propagation, close coupling methods), at Laboratoire Aimé Cotton in Orsay. Such calculations will allow them to describe cold atom?molecule and molecule?molecule collisions, to provide estimates for collisional rates. Their predictions will contribute to guide experimental investigations, and to interpret experimental results, leading to positive experiment?theory feedback.

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Laser-slowing and trapping of molecules for ultracold chemistry

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Objective: In this project, we will investigate collisions between cold homonuclear molecules held in a magneto-optical trap, both theoretically and experimentally. The experiment will be carried out at Prof. Marcassa's group, at University of São Paulo in São Carlos. The main goal in this project is to study collisions in a cold trapped molecular sample in a well-defined quantum state. Such collisions will involve atom-molecule and molecule-molecule collisions in homonuclear samples (Rb-Rb_2 and Rb_2-Rb_2). Dr. Bouloufa's group will carry out molecular structure calculations which will be implemented into various numerical codes for dynamical calculations (grid methods, wave packet propagation, close coupling methods), at Laboratoire Aimé Cotton in Orsay. Such calculations will allow them to describe cold atom-molecule and molecule-molecule collisions, to provide estimates for collisional rates. Their predictions will contribute to guide experimental investigations, and to interpret experimental results, leading to positive experiment-theory feedback.

Abstract: This project was delayed due to the COVID-19 pandemic; however we got the first results in the last year. We were able to perform the first spectroscopical measurements in a cold rovibrational molecular beam.

I. Introduction

In this report, we have focused our attention in producing a well-controlled broadband spectrum and optically pump the molecules to v_X=0. Using such molecular beam, we have performed the first molecular spectroscopy experiments. We have also performed absorption spectroscopy in an Iodine vapor cell, which will be important for our future experiment as a laser calibration tool. Besides, we have characterized our optical cavity.

II. Experiments

II.1. Broadband multimode diode laser source and spectroscopical experiments

Our broadband multimode diode laser system is composed of 7 independent diode lasers in the 680-695 nm range. Each laser provides about 1 W of power. Six diode lasers are combined in a fiber combiner, and they will drive the following $X^1\Sigma^+_g$ (v_x = 1, 2,3,..6) → $B^1\Pi_u$(v_B = 0) transitions. One diode laser is spectrally shaped to drive the $X^1\Sigma^+_g$ (v_x = 0, J_x>10)) → $B^1\Pi_u$(v_B = 1) P and Q transitions. The broadband laser system is shown is Fig.1. In Fig. 2, we show their spectrum. In Fig. 3, we show the shaped spectrum in more detail after de 4F system using a Digital Micromirror Device (DMD).

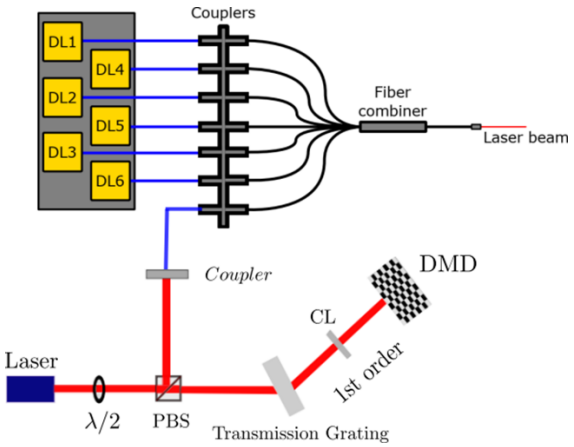

Fig. 1 – Broadband laser system. Six diode lasers are combined in a fiber combiner, and they will drive the $X^1\Sigma^+_g$ (v_x = 1, 2,3,..6) → $B^1\Pi_u$(v_B = 0) transitions. One diode laser is spectrally shaped to drive the $X^1\Sigma^+_g$ (v_x = 0, J_x>10)) → $B^1\Pi_u$(v_B = 1) P and Q transitions.

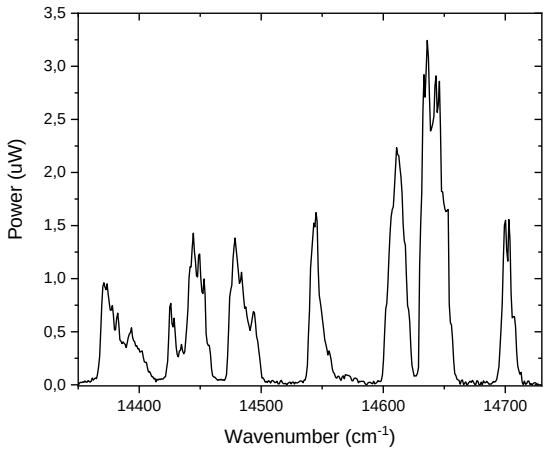

Fig. 2 – Broadband multimode diode laser system spectrum.

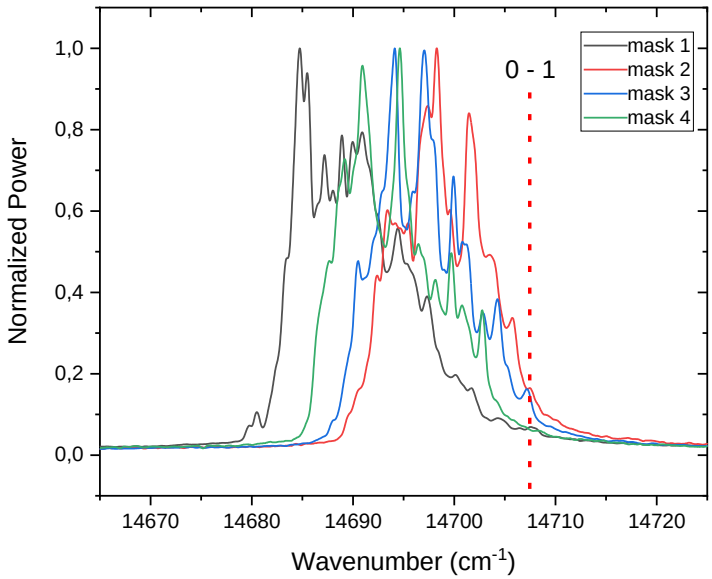

Fig. 3 – Detailed shaped spectrum of the laser to drive the $X^1\Sigma^+_g$ ($v_x = 0$, $J_x>10$)) → $B^1\Pi_u$($v_B = 1$) P and Q transitions, after de 4F system using a Digital Micromirror Device (DMD).

The first experiment, which we performed, was to verify that the rovibrational cooling was working. In this experiment, a photoionization spectroscopy was implemented using two different photons. The first photon, at 680 nm, drives the $X^1\Sigma^+_g(v_x) \rightarrow B^1\Pi_u(v_B = 0)$ transition; and the second photon, at 532 nm, ionizes the Rb_2 molecule. A mass spectrometer allows us to discriminate the Rb_2^+ ion. Fig. 4 shows the involved potentials, and Fig. 5 shows the

spectrum for different conditions of the DMD. Clearly, we can observe rovibrational cooling in the molecular beam.

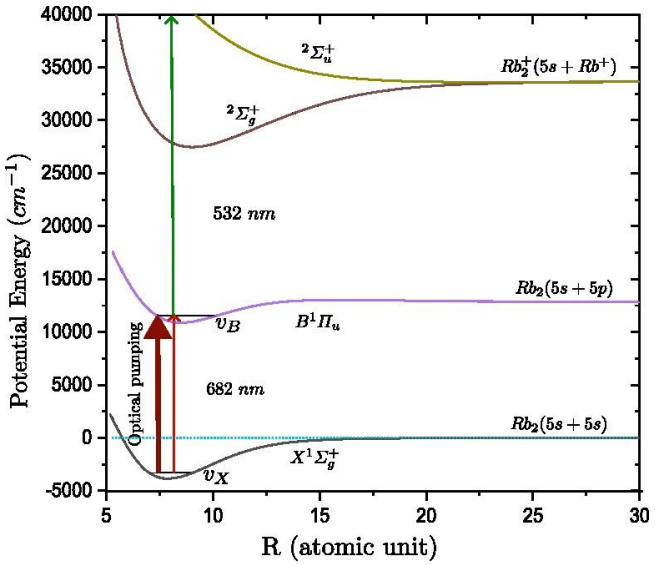

Fig. 4 – Potential curves involved in the photoionization spectroscopy using two pulsed lasers (at 682 and 532 nm). The broadband source was also present to perform the rovibrational cooling.

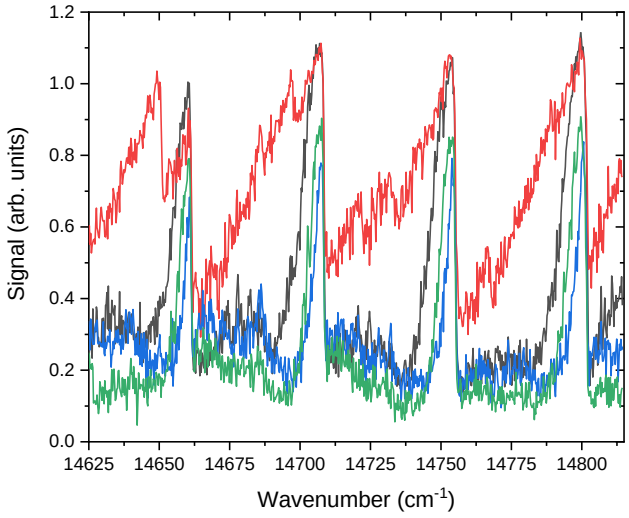

Fig. 5 – Photoionization spectrum as a function of the first photon frequency. The peaks, from left to right, represents $X^1\Sigma^+_g$ (v_x=0) → $B^1\Pi_u$(v_B =0, 1,2,and 3) transitions. Depending on the shaped laser spectrum, the rotational cooling is better implemented. The red curve is the spectrum without the broadband laser source and the others are with different spectral shaping. The narrower the peaks the better is the rotational pumping.

In the second experiment, we have performed photoionization spectroscopy of the $C^1\Pi_u$ potential using a pulsed dye laser around 480 nm, in the coldest rovibrational beam we were able to produce. The first photon drives the $X^1\Sigma^+_g(v_X) \rightarrow C^1\Pi_u$ transition; and the second photon ionizes the Rb_2 molecule. Fig. 6 shows the involved potentials, and Fig. 7 shows the spectrum, which shows $X^1\Sigma^+_g(v_X=0) \rightarrow C^1\Pi_u$ ($v_C=0,\ldots 7$) transitions. Clearly, we can observe rovibrational cooling in the molecular beam. The peak's positions agree with [1], but our theoretical group will improve he theoretical predictions.

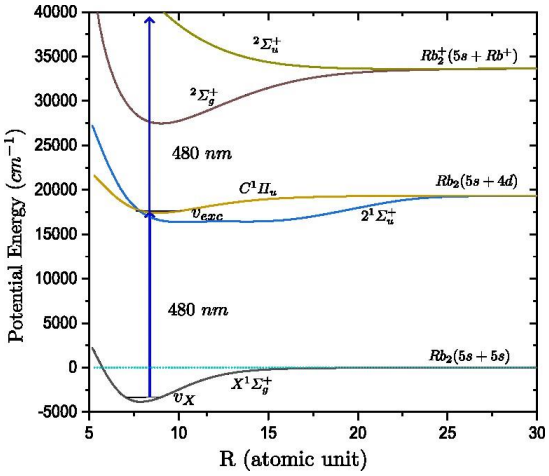

Fig. 6 – Potential curves involved in the photoionization spectroscopy involving the $C^1\Pi_u$ potential using a pulsed laser at 480 nm.

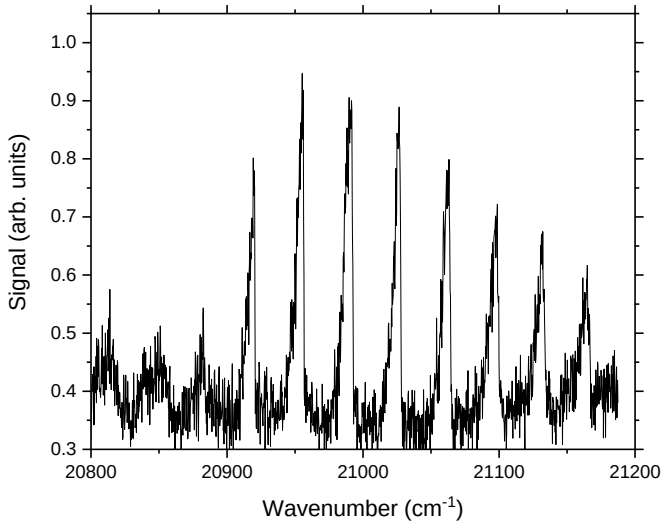

Fig. 7 – Photoionization spectrum shows $X^1\Sigma^+_g(v_X=0) \rightarrow C^1\Pi_u$ ($v_C=0,\ldots 7$) transitions. The peak's positions agree with [1].

In the last experiment, we have performed an experiment to determine the bottom of the $^2\Sigma_g^+$ of the ion molecule in the cold rovibrational beam. Two different pulsed dye lasers were used, one at 682 nm, to drive the transition from the $X^1\Sigma_g^+$ (v_X=0) state to the B $^1\Pi_u$ (v_B=0) state. The other laser was scanned in the 590-600 nm range to drive the transition from the B $^1\Pi_u$ (v_B=0) state to the bottom of the $^2\Sigma_g^+$ (Fig.8). Such potential is important for collisional experiments involving Rb_2^+ [2]. Fig. 9 shows the photoionization spectrum, showing structures consistent with Rydberg contribution and bound states. The LAC team is working to model our results at the moment.

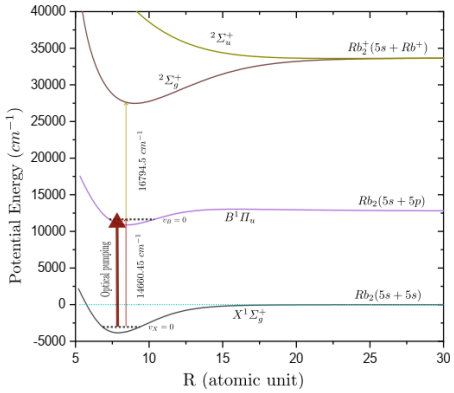

Fig. 8 – Excitation scheme to determine the bottom of the $^2\Sigma_g^+$ of the ion molecule. The first photon drives the transition from the $X^1\Sigma_g^+$ (v_X=0) state to the B $^1\Pi_u$ (v_B=0) state. The second drives the transition from the B $^1\Pi_u$ (v_B=0) state to the bottom of the $^2\Sigma_g^+$.

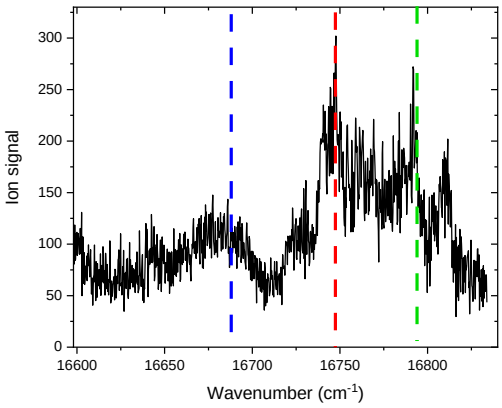

Fig. 9 – Photoionization spectrum show the transitions from B $^1\Pi_u$ (v_B=0) state to the bottom of the $^2\Sigma_g$. The blue line is the measurement from [3], the red line is our limit and the green line the present prediction from LAC's team.

II.2. Optical cavity and Iodine molecule spectrum

We have locked two diode lasers in our homemade optical cavity and performed heterodyne beating. We have measured a linewidth of about 50 kHz after locking them. We have also measured the long drift of the cavity (Fig. 10), which is about 1.2 MHz/day, which

is more than enough to carry out our experiments. We have also performed absorption spectroscopy in a 2 m long I_2 cell at 70^0 C in the 14400-14600 cm^{-1} range, since we have received a new laser in this range. Fig. 11 shows the full obtained spectra, and it is important to point out that our results are better than reference [3].

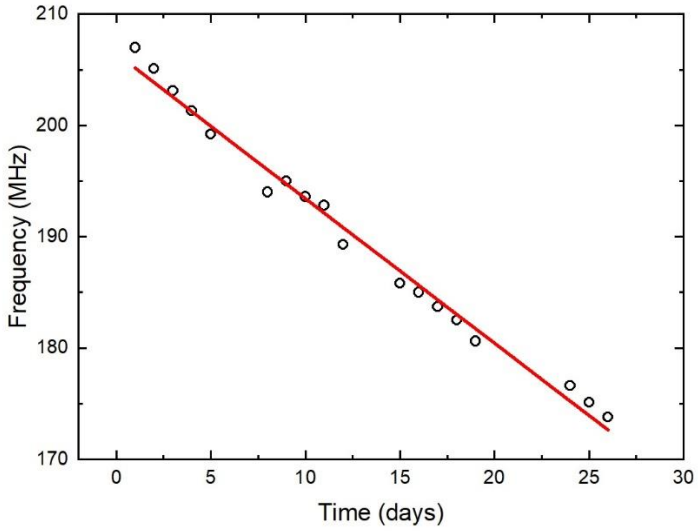

Fig. 10 – Long time drift of the resonance of the optical cavity.

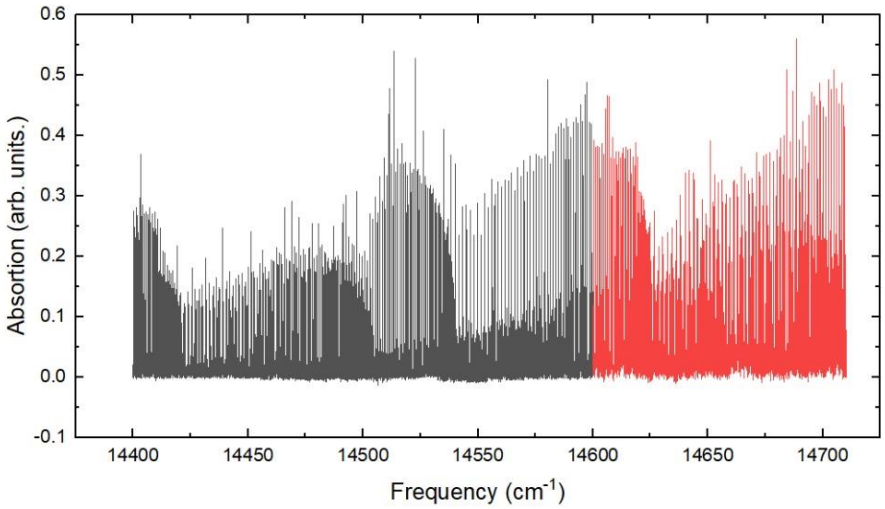

Fig. 11 – Iodine absorption spectra in the 14400-14700 cm^{-1} range. The red part was published in [5].

Personnel Supported

List of personnel associated with the research:

Prof. Dr. Luis Gustavo Marcassa

Dr. Marcos Roberto Cardoso – researcher.

Manuel Alejandro Lefrán Torres – PhD graduate student.

David Rodríguez Fernández - – PhD graduate student.

III. Publications

1. Zeeman slowing of Rb_2 supersonic beam, Manuel Alejandro Lefrán Torres, Henry Fernandes Passagem, Eduardo da Costa Paul, Cristian Adan Mojica Casique and Luis Gustavo Marcassa, J. Phys. B: At. Mol. Opt. Phys. 56, 065301 (2023).

2. High resolution laser spectroscopy of iodine molecule in the 14600-14710 cm^{-1} range, Manuel Alejandro Lefrán Torres, David Rodríguez Fernández, Marcos Roberto Cardoso and Luis Gustavo Marcassa, J. Mol. Spectrosc., 387, 111668 (2022).

3. High resolution laser spectroscopy of iodine molecule in the 14400-14610 cm^{-1} range, Manuel Alejandro Lefrán Torres, David Rodríguez Fernández, Marcos Roberto Cardoso and Luis Gustavo Marcassa, in preparation.

4. Medium finesse optical cavity, David Rodríguez Fernández, Manuel Alejandro Lefrán Torres, Marcos Roberto Cardoso and Luis Gustavo Marcassa, in preparation.

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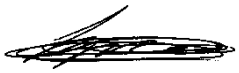
1. J. Amiot, J. Chem. Phys. 93, 8591 (1990).
2. J. Schnabel, et. al, Phys. Rev. A 106, 032804 (2022).
3. M.A. Bellos, et. al., Phys. Rev. A 87, 012508 (2013).
4. H. Salami H. and A. J. Ross, J. Mol. Spectrosc., 233, 157 (2005).
5. M. Lefran et al, J. Mol. Spectrosc., 387, 111668 (2022).

São Carlos, 22/March/2023



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