ATOMIC AND NANOSCOPIC INVESTIGATION USING LASERS (THE LATEST DEVELOPMENTS) HULUMTIME ATOMIKE DHE NANOSKOPIKE ME ANE TE LASEREVE (ZHVILLIME TE FUNDIT)

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PERMBLEDHJE

Ketu propozojme nje menyre te re per matjen e vijave spektral qe nuk munde te maten ne menyre konvencionale. Kemi ndertuar dhe perdoe dy dhomza termike , ne njenen fusim gas, atomet e te cilit i studjojm, kurse ne tjetren fusim edhe atome te elementeve suplementare te cilat sherbejne si shtypje. Eksperimenti qe na formuam me sistemin e detektimit dhe perpunimit te te dhenave bene te mundur matjen e vijave apo kalimeve te ndaluara te cilat sipas informacioneve qe kemi. Na i matim per here te pare. Jane studjuar format e vijave spektrale te Ridbergut nen prezencenm e gazeve suplemtare qe ushtrojne presionin e deshiruart. Cvedosja dhe zgjerimi I vijave spektrale eshte mate ne funksion te shtyypjes dhe te numrit kryesoir kuantik-n.

Fjalet kyce: spektriskopia laserike, dhomzat termike, transicionet

SUMMARY

A new experimental approach is introduced for measurement of spectral lines that can not be measured in a conventional way. Two home made heat pipes are employed. The atoms to be investigated are placed in one of them. The atoms to be studied and atoms used as a pressure are introduced in another pipe. The generated pressure enables one to measure prohibited transition lines that can not be measured otherwise. We have introduces and the experimental setup and required data acquisition system reaching new results. Studies of spectral line shapes of Barium Rydberg states due to rare gas collisions have been conducted. Shifts and broadenings of spectral lines were measured as function of pressure and principal quantum number n.

Key words: laser spectroscopy, heat pipes, transitions

1. INTRODUCTION

Since the availability of tunable lasers, multiphoton absorption techniques have been widely used as standard tools for atomic and molecular spectroscopy. High intensity and narrow line width of lasers provide means to populate Rydberg levels with high principal quantum numbers. It can also produce transitions between energy levels of the same parity commonly known as forbidden transitions in conventional spectroscopy [1-4]. Extensive studies of Rydberg states in alkali atoms and to a lesser extent in the alkaline earth atoms have been reported in the literature. One and two electron atoms provide an important testing ground for theoretical calculations of energy level structure in atomic physics [5-7]. Excited states of alkaline earths (Mg, Ca, Sr and Ba) with two valence electrons exhibit dramatically different features compared to the one electron system of alkalis. Multiple modes of coupling between the two electrons result in configurations which can interact with single electron excited configurations through wave function superpositions[8].

We have started the experimental study of this problem more than one year. While working continuously we have reached only preliminary results. However the new approach we introduced with the home made heat pipes and other experimental elements have constituted a new complex experimental setup for laser spectroscopy. The extreme experimental conditions that can provide this experiment, high temperature variable pressure (with supplementary elements used for the pressure on the vapor of the atoms of elements to be studied, such as Barium), enable one to measure spectral lines in the auto ionized region including called prohibited so transitions.

This setup and approach with some preliminary results presented here we consider that are sufficient for the paper we are presenting. Detailed interpretation of the obtained results (here we present only preliminary results) and further measurement we plane to present in our future papers of the ongoing experiments.

2. METHODOLOGY

Experimental methodology applied was the same as in our previous publications on Barium and Calcium except the addition of few second generation data acquisition systems as shown in Figure1. The high Rydberg states of atoms in a heat pipe are populated by two photon absorption of radiation from tunable dye laser, pumped by high power Excimer laser. Each absorption event produces an ionization signal in the standard thermo-ionic diode which consists of an axial tungsten rod and biased with respect to the walls of the stainless steel cell. The resulting signals are capacitively coupled to an oscilloscope and gated boxcar integrator. Arrangements are provided for the introduction of neutral perturber gases namely He, Ar, Kr and Xe as shown in the block diagram Figure 1. Two home made heat pipes are shown as well.

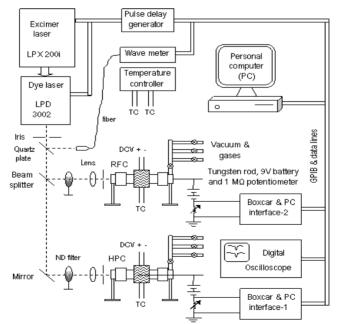


Figure 1. Schematic diagram of the experimental set up. [TC-thermocouple, HPC-high pressure cell, RFC-reference cell, ND-neutral density, DCV-dc voltage and GPIB-general purpose interface bus.

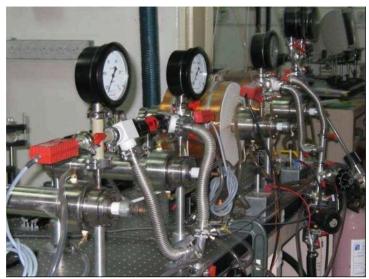


Figure 2. Photograph of experimental set up inside our research lab.

2.1 Experimental

Studies of spectral line shapes of Barium Rydberg states due to rare gas collisions have been conducted. Shifts and broadenings of spectral lines were measured as function of pressure and principal quantum number *n*. We attained very good results comparable with that of other published works. In addition we developed two new stainless steel heat pipes to perform measurements on Calcium, as shown in Figure 2. We have completed the study of doubly excited states of Ba such as 5d6d, 5d7d, 5d7s, 5d8s and 6p² under the influence of perturber gases Ar, Kr and Xe. Furthermore, more experiments were carried out for principal quantum number n = 24to n = 30 to investigate in more detail the effect of perturbing lines on the collisional broadening and shifts of Ba Rydberg states, as one of the most prominent perturbing line 5d7d is lying in this range.

Here we present our preliminary experimental data on the auto-inonized states of Ba that has been carried out. Some of the obtained data are shown in Figures (4-8).The heat pipe ovens are used to produce atomic metal vapor. It consists of an exterior stainless steel tube with water cooling jackets, vacuum connections and fitted with heaters which are capable of heating the cell

up to 1000°C. The ends of the cells are vacuum sealed with guartz windows pressed into O-rings. Water cooling around the ends of the oven cools the O-ring seals, quartz windows and protects the windows from deposition of metal vapor. The main objective is to study the collisional interaction of the alkaline earth Rydberg atoms with rare gases like Ar, Kr and Xe. To study the collisional effect, we set up a reference cell and a high pressure cell. Both cells were heated to approximately 800-810 °C. One of the cells (High Pressure Cell) was filled with high pressure rare gas, while the other cell (Reference Cell) with only 5 mbar Helium as buffer gas. Few grams of Calcium are introduced in the presence of He gas in both cells, before heating. The ion cell detector is biased at +9 V DC with respect to the grounded cell walls. The cells and lenses are positioned so that the laser beam travels parallel to the central tungsten rod and focused at the center of the heated area. The beam travels about 0.4 - 0.6 cm below the rod to prevent any contact of the focused beam with the rod, otherwise data acquisition system may be saturated.

Accurate timing and processing of these signals through gated integrator electronics yields absorption line shape spectra. At every step of the laser wavelength, boxcar averages 30 pulses and the averaged output is fed to the computer interface module (SR 245) and read to the data acquisition computer via GPIB interface. Normally the boxcar gate width was kept at 15 μ s at which optimum signals were collected. Throughout our measurements it is assumed that the resulting

line shapes are identical to Voigt profile. Therefore they were fitted to the Voigt line shape function using Nonlinear Least Squares Fitting algorithm offered in the Origin 7.5 Scientific Graphing and Analysis Software.



Figure 3. Photograph showing experimental cells, vacuum systems, data equipments and the excimer laser (right side orange box).

3. Schematics and image presentation of the experiment:

3.1. Schematic of the present experimental set up with major equipments required.

Equipments required: Excimer laser (LPX 200i), dye laser (LPD 3002), heat pipe oven cells, heaters, chiller, vacuum and gas handling systems, thermocouple monitor, optics & accessories, digital storage oscilloscope, boxcar averagers, pulse generator, computer with GPIB, detector circuit which consists of 9V Battery and 100 k Ω potentiometer.

Experimental setup was upgraded by using the new oscilloscope from Agilent DSO 6104A.

3.2. Pictorial view of experimental setup.

3.3. Pictorial view of our experimental setup with data acquisition system

Complete view of our experimental setup showing four cells, data acquisition equipments including boxcars, oscilloscopes, triggering electronics, wave meter and part of dye laser (orange box to the right).

All technical problems in the experimental setup were rectified in the second year, as mentioned in our second annual report. The availability of DC power supplies, wave meter, a high quality beam splitter and neutral density filters resulted in obtaining noise free and highly reproducible data. Addition of new Digital Storage Oscilloscope (Agilent DSO 6104A) from general facility of science (SAF) presented a major upgrade to our existing data acquisition system.

4. Study of doubly excited states of Ba.

We have been concentrated on the study of collisional shifts and broadening of doubly excited states of barium.

The two cells used for this (reference cell, RFC and high pressure cell, HPC) were the same Ba cells used in the previous year. The inert gases Ar, Kr and Xe were introduced into the HPC to produce collisional broadening of Ba Rydberg states. During the experiment, RFC was pressurized with 5 mbar of He and HPC was pressurized with perturber gases Ar, Kr and Xe of 100 mbar. To cover a complete range of Ba spectrum from the ground state to the ionization limit at $\lambda = 475.79$ nm, we used Coumarin 102, Coumarin 307 and Coumarin 153 dyes. We ran the experiment in the whole range of the spectrum at least three times to check the accuracy and reproducibility of the data.

The broadening and shift of doubly excited states of barium such as 5d6d, 5d7d, 5d7s, 5d8s and $6p^2$ arising from collisions of excited atoms with noble gases Ar, Kr and Xe were measured. We found that configuration interactions due to doubly excited states have strong influence on the regular series, the most pronounced one is from 5d7d configuration near *n*=26. The observed increase in the broadening rate and decrease in the shift rate at n=26 Rydberg state is very surprising. So we examined several states near n=26 to find out the anomalies in the shift and broadening values.

Measured values of the shift and broadening rates of the doubly excited states of Ba are presented in tables and than presented graphically. Typical plots showing the perturbations in the regular series are shown in Figures 4(a-b. The Voigt fitted profile of the 5d7d ${}^{3}F_{2}$ configuration is shown in Figure 6.

4.1. Preliminary results for Auto-ionization of Ba.

Preliminary experiments were carried out on the auto- ionized states of Ba. Detailed study of collisional effects of rare gases on auto-ionized sates of Ba is yet to be started, which is the main focus in our ongoing research. Some of the obtained data are shown in Figures (4-6).

Auto-ionization occurs for all many-electron atoms in highly-excited configurations which lie above the first ionization threshold. Even parity auto-ionized levels of Ba above 6s ionization limit have been investigated by two-photon absorption technique. The 5d state in barium lies above the 6s and below the 6p state. The series originating from these states converge at 5d (2 D_{3/2}, 2 D_{5/2}) and 6p (2 P_{1/2}, 2 P_{3/2}) levels of ionized barium.

Most of the lines in the 5dns and 5dnd series have already been identified.

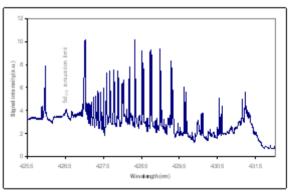


Figure 4. The auto ionizing lines of Ba near the $5d_{3/2}$ ionization limit

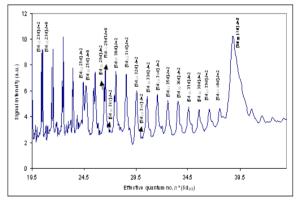


Figure 5. Part of the Figure 4 showing even parity auto-ionizing lines of Ba with effective quantum no n^* near the 5d _{3/2} ionization limit. Wavelength range (426 - 429 nm)

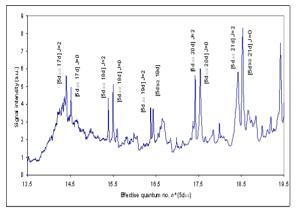


Figure 6. Even parity auto-ionizing lines of Ba with effective quantum no n^* (Wavelength range 429 - 431 nm)

5. CONCLUSION & FUTURE PLANS

We have measured shift and broadening parameters for doubly excited states of Ba and found that configuration interactions have strong influence on the shift and broadening values of the regular series ($6snd^{1,3}D_2$) and the doubly excited states 5d6d, 5d7d, 5d7s, 5d8s and 6p². While these effects have previously been observed in the measurement of radiative lifetimes, absorption cross sections, stark shifts and auto-ionization, it is the first time that their influence on the shift and broadening values is reported. This new finding may be useful for theoretical calculations of interaction potentials and other spectroscopic properties of barium. This could be reached with our new experimental setup and approach. We are expecting new excited and un discovered phenomena to be measured with our experiment.

Our next immediate focus is to complete the Ba and to continue Ca experiments and detailed theoretical studies of the collisional effects of perturber gases on the regular series of Ca. Work pertaining to the auto-ionized lines of Ba above the first ionization limit has been carried out. We have to continue these experiments to measure the shifts and broadenings of these lines under the influence of perturber gas collisions.

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