Proceedings of the Fourth International Workshop on Separation Phenomena in Liquids and Gases

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Preface

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This is the Fourth International Workshop on Separation Phenomena in Liquids and Gases. The Workshop began in 1975 when Professors Bernd and Landahl of Sweden organized the first "Workshop on Gases in Strong Rotation". Because many countries reduced or abandoned their gas centrifuge research programs in favor of the other methods for the isotope separation, the nature of the workshop changed in 1987 when the first "Workshop on Separation Phenomena in Liquids and Gases" was held in Darmstadt. Till 1989 the workshops were held every two years with the understanding that if three years passed with no workshop, then the workshops would be dissolved. Fortunately professor Wood organized very successful "Third International Workshop on Separation Phenomena in Liquids and Gases" in Charlottesville in 1992. Then the workshop stayed alive. The statistics for the ten workshops are shown in Table 1.

It is interesting to see that in 1975 there was only one French participant, but the next workshop was organized in Cadarache, France. In 1983 there was only one Japanese participant, but the next workshop was held in Tokyo. In 1992 there was one Chinese participant but the following workshop was held in Beijing. Again in 1994 we had one Brazilian participant, but the next workshop will be held in Brazil in 1996. The workshops were useful for the scientists in the area of the isotope separation, the mixture separation, etc.

One of the contributions of the workshops was the definition of a hypothetical centrifuge at the "Third Workshop on Gases in Strong Rotation", which was held in Rome. The so called "Rome Centrifuge" allowed scientists to have a "non-classified" set of parameters which could be used for comparing the numerical predictions of the different theoretical models for the flow and separation in the gas centrifuge. The parameters of the Rome centrifuge are listed in table 2.

As for the participant countries you'll find some changes. I was asked at the organizing committee meeting in 1992 to contact with Russians working in the field. You'll see Russia had a large delegation with six members. In the proceedings they presented six papers.

Table 1. Workshop Statistics

	Partici-pants	Host Country	Nations	Papers	Half-day Sessions
'75 Stockholm	37	20	9	22	5
'77 Cadarache	44	16	9	20	4
'79 Rome	70	22	10	28	6
'81 Oxford	70	15	10	26	6
'83 Charlottesville	67	40	10	24	6
'85 Tokyo	90	66	8	22	5
'87 Darmstadt	65	24	13	26	7
'89 Versailles	72	27	15	32	6
'92 Charlottesville	38	13	8	22	6
'94 Beijing	39	20	8	22	6

Table 2. Parameters of the Rome Model Centrifuge

Velocity (m/s)	600
Diameter (m)	0.5
Aspect Ratio Zh/D	10
Length (m)	5
Wall Pressure (torr)	100
Temperature (K)	320

The Fourth Workshop included two laser sessions, three centrifuge sessions, and two sessions about other methods. Twenty-two papers are included in the proceedings. The order of the papers is basically according to that in which they were presented at the workshop. There were eight speeches at the panel discussion. All of the speeches are included in the proceedings.

This Workshop was supported by the National Nature Science Foundation of China and Tsinghua University. Without their supports it would have been very difficult to organize the Workshop. I would like to thank Mr. Yuehui He for his able assistance in organizing the Workshop.

Beijing, December 20,1994

Highlights on the AVLIS program at IEAv/CTA

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Abstract

The aim of the AVLIS program at IEAv is to demonstrate the technical viability of the process using, as long as possible, resources available in Brazil. It implicates not only on studying related processes but also on the development of critical associated technology. We focused our attention mainly on two actuation areas: Copper Vapor and Dye lasers development and spectroscopy. This paper presents some results obtained in our institution in these areas, such as CVL with average output power of up to 40 W and the observation of 3-photon 2-frequency photoionization of ^{238}U both in hollow cathode lamps and in a furnace.

1. Introduction.

The Centro Técnico Aeroespacial, CTA, is a research center that belongs to the Brazilian Ministry of Aeronautics. One of its four institutes is the Instituto de Estudos Avançados (*Advanced Studies Institute*), IEAv, that accomplishes researches in advanced matters that interest, direct or indirectly, that Ministry. The IEAv is divided in six divisions, namely: Nuclear Energy, Theoretical Physics, Experimental Physics, Laser, Technical Support and Computation. The studies related to the Atomic Vapor Laser Isotope Separation (AVLIS) process are performed in the Laser Division, with the support, when required, from the others divisions or even from external institutions. The objective of this program in the IEAv/CTA is to demonstrate the technical viability of the AVLIS process, using as long as possible resources available in Brazil.

This paper presents the activities, related to the AVLIS process, developed in the IEAv, and is mainly focused in two actuation areas, the laser development and the spectroscopy. Some important results, obtained in these areas are also presented.

2. Laser Development.

The AVLIS process needs laser systems that can deliver tunable-in-the-visible beams, with high peak power at high repetition rates. The natural candidate that fills these requirements is the copper vapor laser pumped Dye Laser. In this case, both laser systems work in the Master Oscillator Power Amplifier (MOPA) chain configuration. This section presents briefly the CVL development in our Institute.

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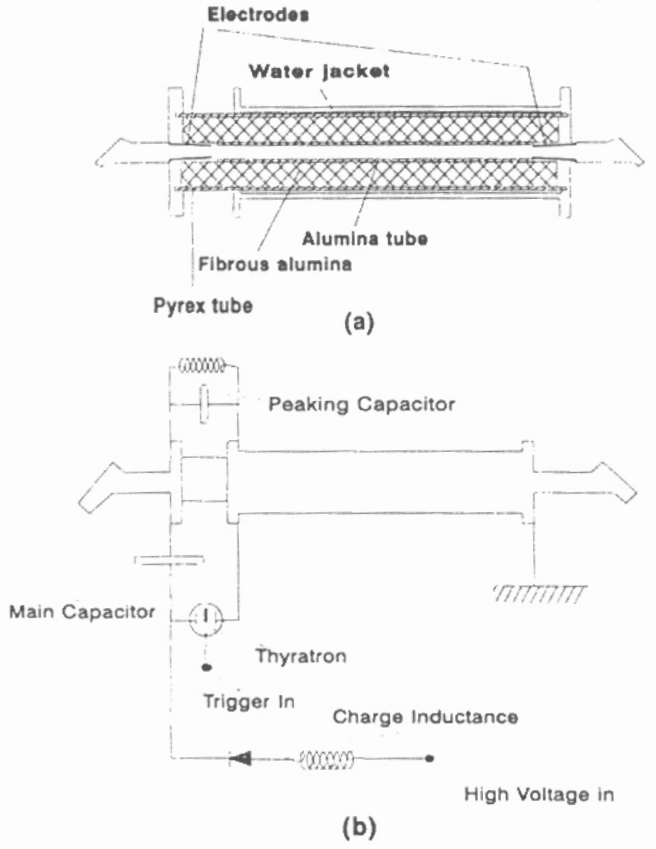


Figure 1: Copper Vapor Laser diagram: a-) laser head; b-) typical excitation circuit.

The CVL development, in the IEAv, started in 1985, with a first prototype of a externally heated copper bromide system that delivered about 100 mW at a repetition rate of 100 pps. From this system the work evolved to self heated true CVLs, with maximum average output power ranging from 5 W, from a compact air cooled system, to 40 W, for a conventional water cooled system.

Every CVL presently in operation in IEAv has the same basic design for the laser head: a high purity alumina tube is surrounded by a thick fibrous alumina thermal insulator and an external Pyrex glass tube assures the vacuum sealing, as shown in Fig. 1-a. There is yet an external concentric water jacket that also provides the coaxial electrical feedback that allows short electric pulses, necessary to the CVL operation. The high purity alumina tube is necessary for it can support high temperatures and, yet, since it has good thermal conductivity, it provides a good temperature uniformity in the gain length. The thermal insulation has to be carefully designed for a given desired output power since the CVL has a narrow temperature range of optimal operation and it is self heated, i.e., the input electric power provides at the same time heating and excitation of the Cu atoms.

There are many different circuits for CVL excitation. One of them, very simple and reliable, used in all CVLs in IEAv, is presented in Fig. 1-b. It is a basic LC inverter circuit where a power supply, with voltage V_0, charges the capacitor C_1 with a voltage

$V \approx 2\ V_0$; when the thyratron is fired, there is a negative pulse ($V \approx -2\ V_0$) and, since the pulse rises very rapidly due to C_2, the energy is preferably transferred to the laser medium. Table I shows the summary of some results obtained with CVL in IEAv [1,2].

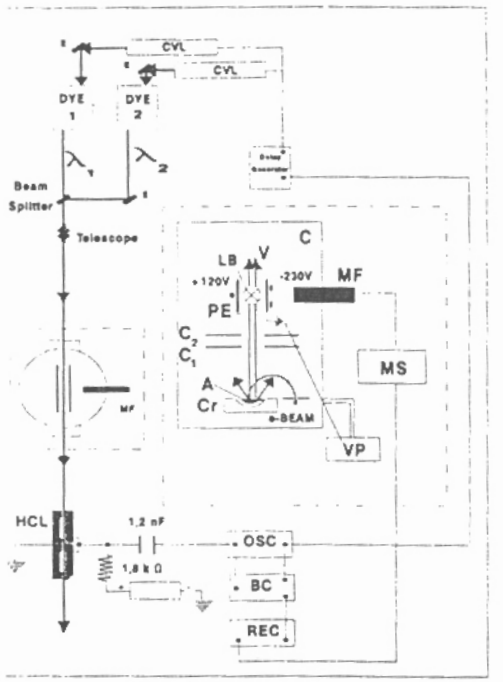
Characteristics	Cu5 - Air	Cu10	Cu40
Average Power (W)	5	11	38
Optimum pulse repetition rate (kHz)	9	9	6
Pulse repetition rate range (kHz)	7-13	6-12	5-10
Green/Yellow ratio (typical)	1.5:1	1.5:1	1.5:1
Pulse energy (mJ)	0.6	1.6	6.5
Pulse width (ns)	30	40	50
Peak power (kW)	20	30	160
Beam diameter (mm)	10	18	42
Full angle divergence (mrad)			
- standard cavity	6	6	6
- unstable resonator	0.5	0.5	0.5
Runtime on one metal load (hours)	60	200	300
Electrical efficiency (%)	0.6	0.92	0.95
Power consumption (kW)	0.84	1.2	4
Gas consumption (liter-atm/hr of neon)	1	1	1
Cooling	Air cooled	2 liter/min tap water (60 psig)	3 liter/min tap water (60 psig)

Table I: Summary of results obtained with CVL development at IEAv/CTA.

CVL development has been continued in IEAv with the study of a new technology, proposed by A. Maitland and C. Little, called HyBrID copper laser [3]. It consists on adding HBr on the gas buffer, such that CuBr is formed in discharge conditions and, later, the salt is dissociated generating the Cu vapor necessary for the laser action. The above mentioned authors argue that with such a technology the operation temperature can be reduced to as low as 400 °C, the efficiency is raised to 3% and, contrary to the conventional CVL, the gain is concentrated at the laser tube axis. Besides the above mentioned advantages, there is still the fact that the big reduction in operation temperature allows the use of cheaper and not so noble materials for the laser heads.

3. Spectroscopy.

Most of the spectroscopy, related to AVLIS process, performed in IEAv/CTA, was accomplished using the set-up diagrammed in Fig. 2. Each tunable dye laser MOPA chain, composed by an oscillator and 2 amplifiers, is pumped by 2 copper vapor lasers. The beams, after combined in beamsplitters, are sent to the furnace and later to the reference hollow cathode lamp, such that the experiments can be done either in the atomic vapor generated in the furnace or in the hollow cathode lamp.



CVL = copper vapor laser MOPA chain
DYE = dye laser MOPA chain
MF = mass filter
MS = mass spectrometer
Cr = crucible
HCL = hollow cathode lamp
V = metal vapor
LB = laser beams
PE = extraction plates
C = vacuum chamber
C_1 and C_2 = thermal ions collectors and collimator slits
A = metal target
OSC = oscilloscope
BC = boxcar averager
REC = xy recorder

Figure 2: Experimental setup.

3.1 Experiments in the hollow cathode lamp.

Let's first comment the experiments performed with the uranium hollow cathode lamp. The first experiment was a two-photon absorption sequence identification. It can be done by interrupting the dye laser #2 beam, varying the dye laser #1 frequency and monitoring the optogalvanic signal. When the laser frequency crosses a resonance, a signal like the solid line shown in Fig. 3 is observed; the laser #1 is tuned to the maximum of the optogalvanic signal and then the laser #2 beam is released and its frequency is varied such that other resonances, due to the second step absorption, are identified, as indicated in Fig. 3. With the set-up described above and with this experimental procedure, we could identify 2 transitions from the ground state and 9 transitions from the 1st excited state. These results are summarized in Fig. 4 [2,4,5]. An interesting remark about the optogalvanic profile obtained for the 5915 Å is that a secondary peak, 3.1 GHz bellow the signal maximum is clearly visible when high intensity is used (we used intensities as high as 200 times the saturation intensity I_s). This secondary peak was attributed to the non-linear two-photon absorption from the ground state to the 33801.03 cm^{-1} level [2,8,11], and this transition plays an important role in the results we are going to analyze ahead.

Another very important parameter to be measured is the absorption cross section. We obtained this parameter for the two mentioned transitions departing from the ground state, using the optogalvanic spectroscopy in hollow cathode lamp and using a very simple

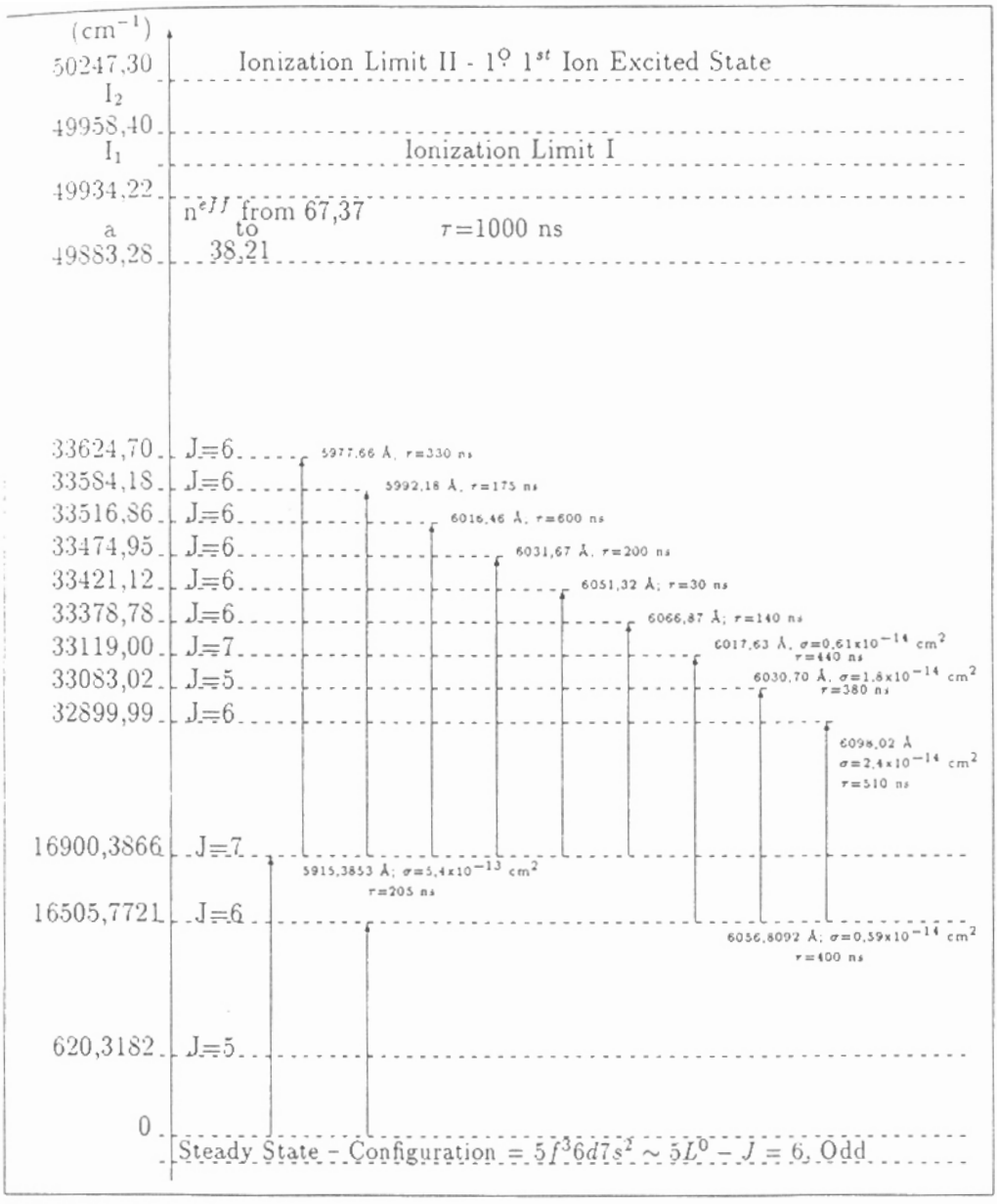


Figure 4: Two photons absorption observed. The lifetime and absorption cross section values are obtained from literature [5-9].

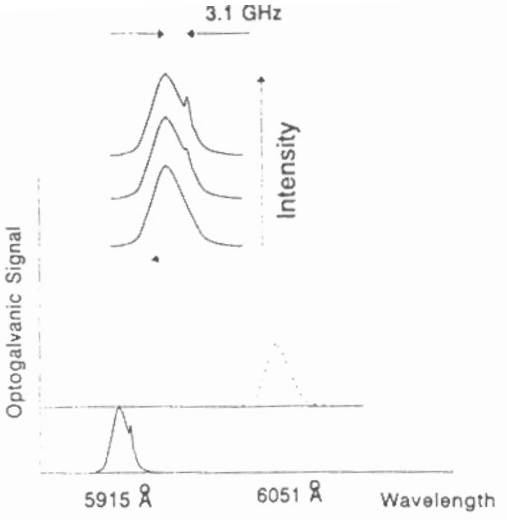


Figure 3: Diagram of the optogalvanic signal for two-photon absorption sequence identification. The detail indicates the secondary peak amplitude dependence with the probe beam intensity.

model to describe the optogalvanic signal, that takes into account the non-homogeneous broadening nature of the absorption lines (characteristic for absorption media generated in hollow cathode lamps).

The first assumption in our model is that only the population of the levels participating on the absorption process, denoted by n_1 and n_2, are significatively changed and that the optogalvanic signal is proportional to this change [12,13], or rather,

$$\Delta V \propto \Delta n_2 \tag{1}$$

where Δn_2 is the upper level population density fluctuation due to light absorption.

The rate equation for a homogeneously broadened two-level system interacting with a resonant light beam is given by [14]

$$\frac{dN}{dt} = -\left(1+\frac{I}{I_s}\right)\frac{N}{\tau} + \frac{N_0}{\tau} \tag{2}$$

where

$$N = n_1 - n_2,$$

$$I_s = \frac{h\nu}{\sigma\tau},$$

$$N_0 = N(I=0).$$

Then the second assumption is made: the absorbing medium in a hollow cathode lamp is characteristically Doppler broadened and to take this aspect into consideration we introduced a *power broadening factor* substituting N by $N/\sqrt{1+I/I_s}$ in Eq. 2 that becomes

$$\frac{dN}{dt} = -\left(1+\frac{I}{I_s}\right)^{1/2}\frac{N}{\tau} + \frac{N_0}{\tau}. \tag{3}$$

If we consider yet that only a small fraction of the laser beam is absorbed, i.e., $\Delta I << I$, the solution for Eq. 3 is

$$\Delta V = A\left\{1 - \frac{1}{\sqrt{1+I/I_s}}\left[1+\left(\sqrt{1+I/I_s}-1\right)e^{-\sqrt{1+I/I_s}\,t/\tau}\right]\right\}. \tag{4}$$

If one measures the optogalvanic signal varying the income beam intensity and fit Eq. 4 to these experimental results, one obtains I_s and, knowing the relaxation time for each level, the absorption cross sections.

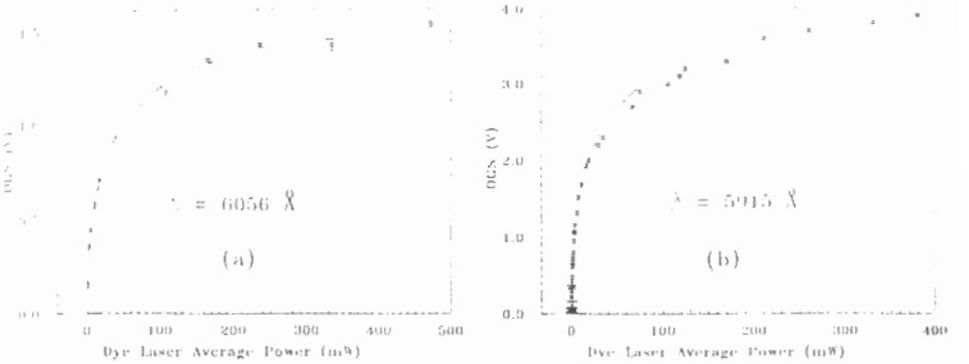


Figure 5: Optogalvanic signal against probe beam input average power. The symbols indicate the experimental values and the line the best fit for Eq. 4.

Fig. 5-a shows the experimental results for the optogalvanic signal against the beam intensity for the 6056 Å transition. The solid line is the best fit of Eq. 4 for these data and the obtained absorption cross section is $\sigma = 1.2\times10^{-14}$ cm^2, amazingly close to the results presented in the literature of $\sigma = 0.6\times10^{-14}$ cm^2 [7]. However, when this method was tried with the 5915 Å transition (Fig. 5-b), not only the cross section is very far from those met in the literature, but also its value depends on what experimental values we use to fit Eq. 4, that's to say, the higher the input intensity, the higher the I_s given by the best fit. Actually, these results should be expected, since, as said before, this transition has, very close to it, a non-linear two-photon transition to the 33801.03 cm^{-1}. When the input intensity increases, not only the two-photon transition probability increases but also, due to the power broadening, the transition from the 16900.39 cm^{-1} (5915 Å) level to the 33801.03 cm^{-1} is more and more important. Thus, if this explanation is valid, a trend analyses should indicate, for very low value of input intensities, the true *Is* value of the 5915 Å transition. Fig. 6 shows the fitted I_s values against the maximum input intensity used for the fitting. If one fits Eq. 4 using all the experimental values shown in Fig. 5-b, one gets the last point indicated in the Fig. 6; if one takes all the points except the last one, one gets the point before the last one; if one eliminates the two last experimental points in the fitting, one gets the point before the two last ones, and so on. The solid line is just an eye fitted curve to indicate the trend. If we take the value of $I_s \approx 0.18$ W/cm^2 as the actual saturation intensity for that transition, we obtain $\sigma = 4.9\times10^{-13}$ cm^2 , what is once again very close the results found in the literature of $\sigma = 5.4\times10^{-13}$ cm^2 [9]. We don't claim, however, that these results confirm the assumption taken in our simple model, but it seems to us that this method can be used to obtain experimentally very reasonable estimation of saturation intensities.

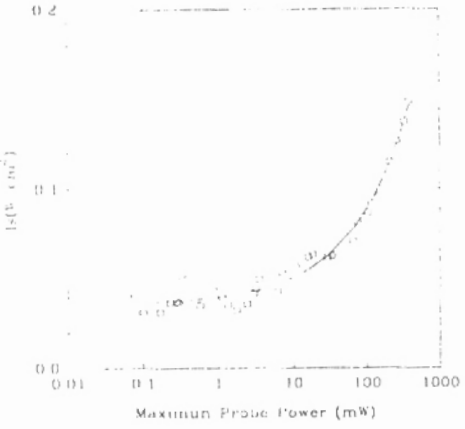


Figure 6: Saturation intensity obtained by fitting Eq. 4 to the experimental data shown in Fig. 5-b against the input power of the last point taken for the fitting. Averaging I_S, for input power smaller than 1 mW, we obtained $I_S \approx 0.18$ W/cm^2

3.2 Experiments in the furnace.

Although the two-photon scheme is far from the ideal for the AVLIS, some ionization experiments were performed with interesting results [2]. These experiments consists on three steps: i-) with the laser beams interrupted, the mass spectrometer ionization filament is turned on and the mass spectrum for the evaporated U is taken, showing the characteristic natural isotopic abundance of 99.3% of ^{238}U and 0.7% of ^{235}U; ii-) with the filament off, the laser beams are released and tuned in the 5915 Å and 6051 Å, monitoring the optogalvanic signal, as described in section 3.1, the mass spectrum is taken and only the profile due to the ^{238}U appears, showing good selective ionization of this isotope; iii-) the laser #1 is detuned 3.1 GHz toward the non-linear two-photon absorption pointed in section 3.1, the ^{238}U mass spectrometer is still very clear but with a magnitude 20 times bigger than that one described in step (ii). Fig. 7 shows the simultaneous optogalvanic and the ^{238}U photoionization spectra when the laser #1 frequency is scanned; it is clear there the 3.1 GHz detuning between spectrum maxima. These results are understood as follows. In the step (i) the mass spectrum is just that one for natural U non-selectively ionized, showing thus the natural concentration of the main isotopes. In the step (ii) four different processes can contribute to the ion signal, as diagrammed in the left side of Fig. 8: a-) the first frequency excites the atom to the 16900 cm^{-1} level, the second frequency excites the atom to the 33421 cm^{-1} level and the first frequency ionizes the atom; b-) the same as in (a) but with the ionization promoted by the second frequency; c-) the first frequency excites the atom to 16900 cm^{-1} and, due to power broadening, the same frequency excites the atom to the 33801 cm^{-1} level and, finally, the second frequency ionizes the atom; d-) the same as in (c) but the with the ionization also due to the first frequency.

In step (iii) a two-photon absorption excites the atom to the 33801 cm^{-1} level and than the atom can be ionized either by the first or the second frequency, as indicated in the right side of Fig. 8.

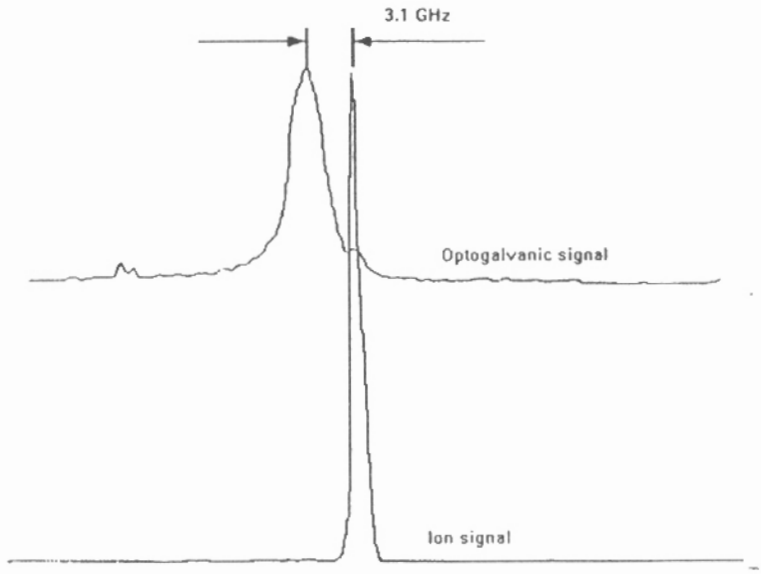


Figure 7: Optogalvanic and photoionization signal, for two wavelengths excitation ($\lambda_1 \approx 5915$ Å, $\lambda_2 = 6051$Å). Laser #1 wavelength is scanned near the resonance. The ion signal spectra is understood as been due to a three-photon two-frequency photoionization process.

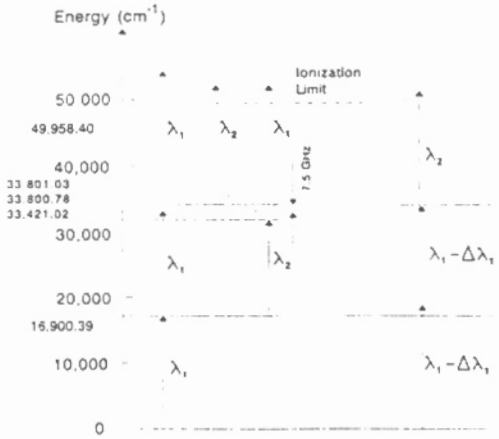


Figure 8: Diagram of the possible mechanisms for U photoionization, using two-frequency three-photon absorption with $\lambda_1 \approx 5915$ Å and $\lambda_2 \approx 6051$ Å. The left side indicates the situation when λ_1 is coincident to the absorption peak and the right side indicates when λ_1 is detuned 3.1 GHz, as indicated in Fig. 7.

4. Conclusions

This paper presented some aspects of the AVLIS program at IEAv/CTA. One of the main goal, in this program, is the achievement of the CVL construction technology, where we obtained lasers with characteristics close to the commercial systems, available in the international market with severe restrictions by the side of the exporter country governments.

In the field of the spectroscopy, although a practical three frequencies sequence have not been yet identified, we have already obtained the necessary experience to start the investigation of the multi-frequency absorption in the atom of uranium, in order to get convenient line sequences for the AVLIS process. Since we started to work in the third laser system, to generate the third frequency, we hope to obtain very soon these desired data.

5. Acknowledgments

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