

URANIUM ISOTOPIC ENRICHMENT USING LASERS

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RESUMO

Vários países que utilizam energia nuclear têm investido significativamente no desenvolvimento de técnicas a laser aplicadas na separação de isótopos. No Brasil, estudos e experimentos sobre separação de isótopos de urânio por lasers vêm sendo desenvolvidos no Instituto de Estudos Avançados (IEAv) do Centro Técnico Aeroespacial (CTA). Estes estudos têm por objetivo a demonstração da viabilidade deste processo usando, sempre que possível, os recursos disponíveis no País. Neste trabalho descrevemos resumidamente o método de separação isotópica a laser e apresentamos o presente status de Pesquisa e Desenvolvimento nesta área. Também descrevemos alguns resultados obtidos nos nossos laboratórios. Damos ênfase, neste trabalho, ao processo atômico de separação de isótopos a laser.

ABSTRACT

Several countries that utilize nuclear energy are investing significantly in the development of laser techniques applied to isotope separation. In Brazil these studies are concentrated in one research institute, the IEAv (Institute for Advanced Studies), and aim towards demonstrating the viability of this process using, as much as possible, resources available in the country. In this paper we briefly describe the laser methods for isotope separation, giving an overview of the present research and development status in this area. We also show some results obtained in our laboratories. We focused this report on the atomic route for laser isotope separation.

1. INTRODUCTION

The most used element as fuel for nuclear reactors is uranium, whose natural composition consists on 99.275% of the ^{238}U isotope; 0.720% of the ^{235}U isotope and 0.005% of the ^{234}U isotope. The great majority of the existing nuclear reactors, on the other hand, requires previously enriched uranium, i.e., the uranium must have its contents of ^{235}U isotope increased to a concentration between 3 - 4%. The isotope enrichment is the most difficult stage in the whole process of producing the nuclear fuel. As isotopes of one

element are chemically identical, they should be separated by methods based on their physical properties. The traditional methods for uranium enrichment are gaseous diffusion and ultracentrifugation. These processes are based in the small mass differences between the isotopes. The enrichment factors (the relation between the concentrations of product and of feed materials) obtained in one single stage lay between 1.004 (for gaseous diffusion) and 1.3 (for ultracentrifugation), so that, in order to achieve the necessary enrichment to be used in the reactors, it is necessary to proceed through hundreds of successive stages, the so called *cascaes* of enrichment.

In this paper we discuss some aspects of a technology, proposed in the seventies to compete with the traditional ones: the laser methods for isotope separation (LIS). We present a summary of the status of research and development in this area around the world, briefly describe how our group started to work on this subject, discuss the physical principles of the laser methods and present some highlights on the Atomic Vapor Laser Isotope Separation (AVLIS) at IEAv.

2. PRESENT STATUS OF LASER ISOTOPE SEPARATION DEVELOPMENT

Many countries, among those which utilize nuclear energy around the world, have significantly invested, in the last twenty years, in research and development of new technologies for producing enrichment of uranium. The traditional methods for producing nuclear fuel are probably going to be gradually abandoned, in favor of these new technologies, and the main reason for this is the perspective of a drastic reduction in the costs of production of enriched uranium.

Table 1 shows a summary of the status of research and development in uranium isotope enrichment around the world.

The development of new structural materials, such as Al, Ti and other special metal alloys and composites achieved mainly by the aerospace industry, in the last 40 years, made the ultracentrifugation method to reach an extreme level of efficiency, so that nowadays ultracentrifugation and the laser separation methods seem to be the most competitive among the separation processes. The first one is already used in an industrial scale by URENCO. The second one is, in most of the interested countries, in a stage of research and development. Nevertheless, recent

statements [1 ; 2] from the *U. S. Enrichment Corp. (USEC)* and from the *U. S. Department of Energy (US-DoE)* concerning the purpose of operating a commercial AVLIS facility in the very beginning of the next century shall lead to an increase of the investments in other countries to implement similar projects.

The U. S. choice by the AVLIS process was due mainly to economic factors [3]. Over and above that, the modular nature of AVLIS allows the installation of smaller plants, which can be easily upgraded according to the fuel demand [4], while the other processes demand always large

scale plants, due to their smaller separation factors. AVLIS looks, therefore, the most indicated process to substitute the traditional ones in producing enriched uranium. Nevertheless, a successful commercialization of this technology will threaten well established fuel cycle activities. Different specifications of feed and products and the necessary changes in the interfaces may generate resistance to the deployment of the AVLIS technology from users and from companies in the fuel cycle activities. In spite of this, several countries proceed with plans and studies on the new laser processes.

Table 1 - Research and development in uranium laser isotopic enrichment. This table was taken from Cavalcante [5] and updated with data taken from [1, 6, 7].

COUNTRY	DIFFUSION	CENTRIF	CHEMICAL	HELIKON	JET NOZZLE	LASER	PLASMA
South Africa				A		B	
Argentina	A						
Australia		B				B	
Brazil		A			A ^{h*}	B	D
Spain	B ^b , D						
U.S.A	C	A ^{**}	B			A, F	B
France	C	B	A			A	B
Holland		C ^c				B	
India ^{**}							
Israel						B	
Italy	B	B				B	
Yugoslavian		P _A	P _B			P _B	
Japan	B, B ^c	C	A			B	
Pakistan	P ^f	C, C ^e					
Germany		C ^c			A ^d	B	
China	C	C ^e				B	
Sweden						B	
Switzerland			B			B	
U.K.		C ^b				A	
USSR	C	E					

Stage of R&D	
A -	Pre-industrial
B -	Laboratory/ Pilot
C -	Industrial
D -	Research
E -	Lack of information
F -	Technology ready for industrialization
P	Project
P _A	Industrial project
P _B	Laboratory project

Cooperation	
a -	Brazil/Germany
b -	EURODIF
c -	URENCO
d -	Japan/USA
e -	Pakistan/China
f -	Pakistan/Argentina

U.S.A	United States of America.
U.K.	United Kingdom
USSR	Former Soviet Union

- * Suspended activities
- ** Agreement with Russia (Unknown method)

3. HISTORY OF THE BRAZILIAN LASER ISOTOPE SEPARATION PROGRAM

In the year 1972 the USA started the laser isotope separation program in Los Alamos [8] and in Lawrence Livermore [9]. In this same year Sergio Porto, a Brazilian researcher, was a professor at the Caltech and in the

University of South California, and had some meetings ("social meetings") with Brazilian students in the USA. They had discussed the possibility of having a similar laser program when they returned to Brazil. One of these students, an officer of the Brazilian Air Force called José A. A. Amarante, founded effectively a laser group as part of what was later called the Brazilian autonomous nuclear program.

In 1974 an agreement between the National Nuclear Energy Commission (CNEN) and the Aerospace Technical Center (CTA) created the isotope separation program. In this same year it was also signed a research contract between CTA and the State University of Campinas (UNICAMP). Some very promising issues related to laser isotope separation in nitrogen and in boron resulted from this cooperation. Although, the real interest of these studies was the uranium enrichment. In this respect the progress was not so quick. After some preliminary experiments in the molecular route at UNICAMP and in the atomic route at the Institute for Atomic Energy (IEA), a new research institute was founded, in the city of São José dos Campos, in the state of São Paulo. Since then experiments on laser isotope separation have been concentrated at the Institute for Advanced Studies (IEAv).

4. PHYSICAL PRINCIPLES OF THE LIS METHODS

The physical principles of the laser methods for uranium isotope separation differ fundamentally from traditional methods. In the case of ultracentrifugation and gaseous diffusion, the separation is based on mass differences between the various isotopes. The fundamental parameter in the laser methods is the differentiated capacities possessed by the different isotopes of absorbing light of particular frequencies.

The various ways of separation by laser differ on the way how the selective excitation is attained and by how this excitation is transformed into separation. According to this one can make distinction between the two routes for laser isotope separation: the molecular and the atomic routes. In the molecular route, the energies of interest are those of the molecular vibrations, which present shifts in the infrared absorption spectra. These isotopic shifts are proportional to the reduced masses of the vibrators. The isotopic shifts, in spite of the fact of being relatively large, are usually, and specially for the heavier elements, hidden by overlapping due to hot bands and rotational structures [10]. The isotopic structures can, in such cases, only be revealed if the molecules are cooled down to about 50 K. In the case of uranium separation, the molecules of UF_6 are diluted with a carrier gas and adiabatically expanded into the vacuum, in order to prevent condensation by cooling. UF_6 can then be selectively excited using $16 \mu\text{m}$ lasers (resonant with the ν_3 vibration at 630 cm^{-1}) and undergo photolysis to UF_5 , which is solid and easily separated from the gas phase.

In atoms, the isotopic differences in the absorption spectra are caused by the volume effect of the nucleus and by the nuclear spin of the isotopes. The electronic energy levels present shifts in the visible range of the spectrum, and such shifts allow one of the isotopes to be selectively excited by a monochromatic laser beam. In the case of uranium, the two principal isotopic species differ by 3 atomic mass units in their nuclei. This is a very small difference, if compared to the large nuclear masses.

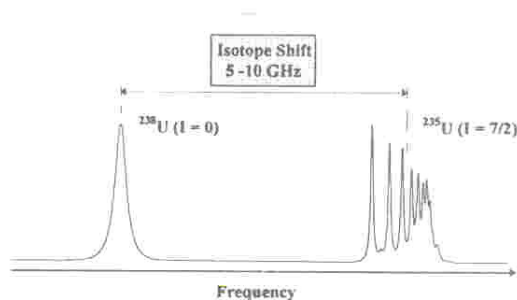


Figure 1: Appearance of the absorption spectrum of uranium around 600 nm.

The appearance of a spectrum of uranium at wavelengths around 600 nm can be seen in Figure 1. Of course, the scale was changed between the two absorption lines. Two effects can be observed here: a 5-10 GHz frequency shift and a difference in the line structure. The last one is the hyperfine structure due to the nuclear spin of the ^{235}U isotope. There are actually 21 lines which are not resolved in this figure [11].

Thus, if a mixture of these two isotopes is irradiated by a laser which has a resonant frequency with the ^{235}U isotope and a sufficiently narrow line, the light of the laser will be preferentially absorbed by this isotope.

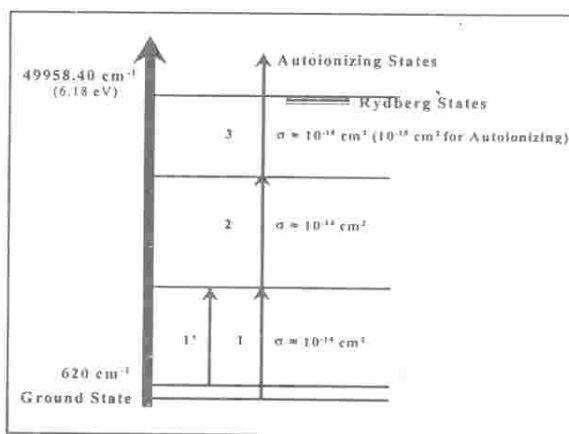


Figure 2: Three-photon, four-color scheme for uranium photoionization.

The isotopic separation of uranium by laser and metal vapor (AVLIS) is based on this fact. First, metallic uranium is vaporized by an electron beam gun. The ^{235}U isotope present in this vapor is selectively excited by laser. This selective laser absorption step is represented by the line 1 of Figure 2. In order to be ionized, the atoms must absorb other photons so that the total energy of these photons together is greater than the ionization limit (6.18 eV). This corresponds to the transitions 2 and 3 of Figure 2. None of these frequencies is resonant with the ^{238}U atoms, which are, therefore, not affected during the process. This Figure shows what seems to be the most efficient way for selectively excite and ionize uranium atoms [12]. It is a three-photon, four-colors scheme, where the fourth color is to be used at any practical plant,

considering that, in normal conditions of the process, around 30% of the atoms in the vapor are in the first excited state at 620 cm^{-1} .

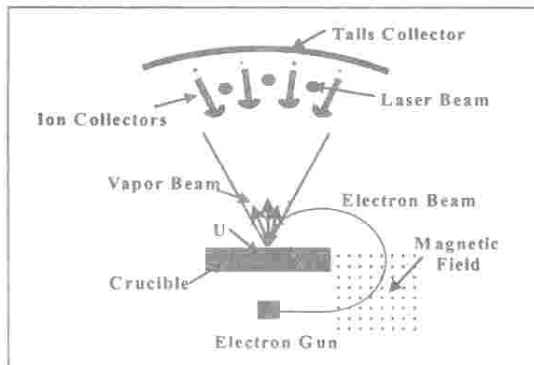


Figure 3: Scheme of the evaporation and collection system.

After the photoionization, the ions can be deflected by electrical and/or magnetic fields and guided up to a collector located in a place not accessible to the neutral ^{238}U atoms, as shown in Figure 3.

4.1 Some Aspects of the Molecular Route

Photochemical studies are presently being carried out on a variety of compounds and on their decomposition products, in order to obtain a method of deuterium separation with CO_2 lasers. A study on the isotopic exchange between H and D was initiated with this objective.

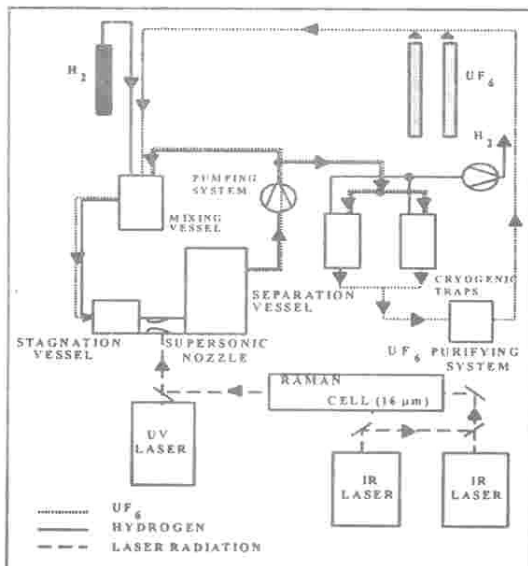


Figure 4: Experimental setup for molecular uranium laser isotope separation.

Another interesting subject under study at IEAv aiming towards isotopic separation by lasers in the molecular process combines the formation of molecular clusters by a gasdynamical process and the inhibition or

destruction of these clusters in one of the isotopic variations by means of laser radiation [13, 14].

With respect to uranium, Figure 4 shows the sketch of the experimental apparatus used at our laboratory. UF_6 is expanded together with hydrogen in a supersonic nozzle. The $16\ \mu\text{m}$ radiation is produced by Raman conversion of TEA CO_2 lasers in a multipass para-hydrogen cell. This radiation excites selectively the $^{235}\text{UF}_6$ molecules. Photolysis is then performed by UV lasers.

4.2 Some Aspects of the Atomic Route

A basic unit used in the AVLIS process is formed by two mechanically independent systems. In what we call the "evaporation system" occur the processes of atomic vapor generation, interaction between atoms and light, collection of ^{235}U ions, and of depleted uranium. This system includes the electron gun, the vacuum system, the crucible with metallic uranium, mass spectrometer and other probe systems for the vapor and for the generated plasma, and the product and tails collectors.

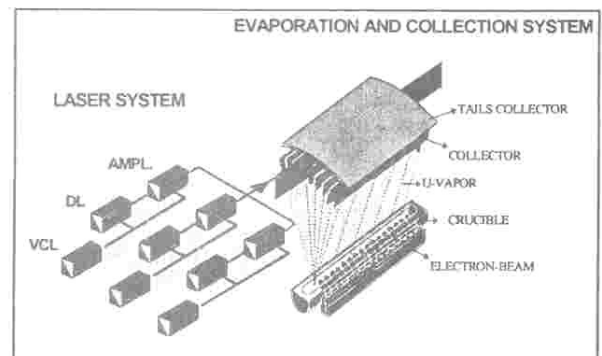


Figure 5: AVLIS basic unit.

In the "laser system", on the other hand, occurs the production of the necessary radiation for the photoexcitation and photoionization of the selected isotope. It includes basically the copper vapor and dye lasers, the systems of power and tuning control of the lasers and the beam combination and transport optics.

All the processes mentioned above are complex and their studies are very important, because their knowledge offers the many possibilities of selecting and combining parameters that determine the performance of the system.

Figure 5 gives an idea of the basic module as described before. This scheme is obviously very simplified here. In an industrial plant uranium must be fed continuously and also continuously collected. In a laboratory-scale experiment, such as our experiments, uranium is fed once as an ingot and, after evaporation and photoionization, the product is collected in a cold plate, where the vapor is cooled down to solid state. Then the system is opened and after crucible and collector replacement, a new run begins. The metallic uranium contained in the crucible is evaporated by an electron beam. The vapor expands upwards in vacuum and is then

illuminated by the laser system and selectively ionized. The ^{235}U Uranium ions are extracted from the neutral background by means of suitable fields and directed towards the collectors, while the depleted vapor flows to the respective tails collector. These processes occur inside a vacuum chamber that must offer some facilities for system alignment, handling, and cleaning, as the processed material is toxic.

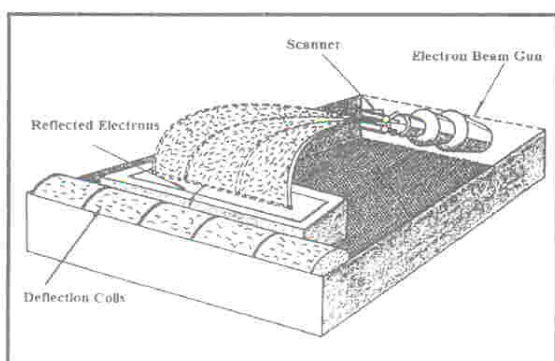


Figure 6: Fast scanned, point electron gun.

Uranium is very corrosive at high temperatures. That is why the cathode of the e-gun must be kept protected from the atomic vapor. Otherwise, the uranium vapor condensed on the e-beam gun filament will produce an alloy that degrades quickly its performance. Then the filament should not have a direct view of the crucible and the beam of electrons must be deflected by magnetic fields, as shown in Figure 3 and Figure 5. The deflecting and the conforming fields must be subject of careful calculations, specially taking into account that in the case of high power guns phenomena caused by spatial charges have considerable effects.

The use of electron beams presents some advantages over the Joule heating. First, it reduces the effects of corrosion. In the case of Joule heating, heat is transferred to the crucible, whose interface with the liquid uranium is at high temperatures. With the use of an e-beam the heating is localized on the free surface. The interface between the uranium and the crucible can even be maintained in the solid state. Secondly, the e-beam allows to achieve higher vapor densities in the interaction region and consumes less energy than the Joule heating.

Uranium is vaporized by means of a high energy electron gun. The metal vapor must be confined in a region with a suitable shape, in order to maximize the interaction with the laser beam. It means small cross section and a long length which must be crossed longitudinally by the laser. This effect can be obtained in two different ways: by a linear electron beam or by a point electron beam driven by a fast scanning system. In the first case, the electrons are emitted from a long filament and after being properly shaped by suitable fields are guided and focused onto a linear region on the surface of the sample. In the second case a conventional electron gun is used, whose focus is nearly punctual, and then a fast scan is made in one

dimension, as shown in Figure 6. In both cases the vapor is produced in a nearly linear region and expands upwards to the interaction region.

In the interaction region the continuously flowing vapor must be illuminated by high power pulsed lasers. Beside the usual laser properties, such as tunability, high intensity, and coherence, the lasers used must possess a high repetition rate in order to illuminate most of the atoms. In order to preserve the selectivity of the excitation, it is also necessary to have a pulse duration smaller than the particle collision time ($\sim 10^{-6}$ s) and smaller than the radioactive relaxation time of the intermediate state ($\sim 10^{-7}$ s). The best choice presently is represented by tunable pulsed dye lasers pumped by copper vapor lasers. Copper vapor lasers show efficiencies of about 1%, power up to 200 W, and up to 10 kHz repetition rates.

The photoionization process presents a very high selectivity. It is about 100% in our experiments. Nevertheless, the overall selectivity is degraded in the process of ion extraction and collection, so that the assay of the product mixture lies usually between 5% and 60% of ^{235}U .

The enrichment factor of an isotopic separation process is defined as the relation between the isotopic ratios in the product and in the feed ($\alpha = R_p/R_f$) in one stage. In the process we are describing (AVLIS), this quantity lies between 7 and 200. These values are much higher than those reached by each stage of gaseous diffusion ($\alpha=1.002$) and ultracentrifugation ($\alpha < 1.3$) methods. This fact allows the necessary enrichment of uranium for nuclear reactors in only one stage. Moreover, it allows the production of enriched uranium from materials already depleted by the traditional means ("tails stripping").

5. HIGHLIGHTS ON THE AVLIS AT IEAV/CTA

In order to cover all the steps of the process, extensive studies are presently being carried out on subjects such as:

1. Vapor production including electron-beam technology, studies on the uranium-crucible system (corrosion, buoyancy and Marangoni flows) and on the vapor expansion.

One of the main technological problem in the AVLIS process is the generation of metallic uranium vapor. Due to the intrinsic characteristics of the process, this vapor must be dense and contain only neutral uranium atoms in a very directional beam. High densities are necessary since large amounts of enriched uranium at high production rates are desired; charge neutrality is necessary otherwise all ions present on the laser-vapor interaction region will be attracted to the collectors, decreasing the selectivity obtained by lasers; and the vapor beam must be directional because, making the laser beams perpendicular to the vapor beam, Doppler spectral broadening can be avoided. Additionally, these three aspects are dependent from each other.

Metal evaporation for AVLIS purposes is a multivariable problem and the trade off among these variables is a hard task to accomplish. Our first priority was the spectroscopy and the laser development aspects, however some studies about uranium evaporation have been performed. A numerical code to describe the plasma behavior and the collection mechanisms was developed and a study of special refractory materials for crucibles that are resistant to uranium attack is being accomplished.

Table 2 - Summary of CVL Achievements.

Characteristics	Cu5 - Air	Cu10	Cu40	Cu-HBr
Average Power (W)	5	11	38	27
Optimum pulse repetition rate (kHz)	9	9	6	18
Pulse repetition rate range (kHz)	7-13	6-12	5-10	8-20
Green/Yellow ratio (typical)	1.5:1	1.5:1	1.5:1	1.3:1
Pulse energy (mJ)	0.6	1.6	6.5	1.5
Pulse width (ns)	30	40	50	30
Peak power (kW)	20	30	160	50
Beam diameter (mm)	10	18	42	15
Fill angle divergence (mrad-standard cavity - unstable resonator)	6 0.5	6 0.5	6 0.5	N.A.
Runtime on one metal load (hours)	60	200	300	N.A.
Electrical efficiency (%)	0.6	0.92	0.95	1.3
Power consumption (kW)	0.84	1.2	4	2.1
Gas consumption (liter-atm/hr of neon)	1	1	1	N.A.
Cooling	Air cooled	2 liter/min tap water (60 psig)	3 liter/min tap water (60 psig)	1 liter/min tap water (60 psig)

2. Photon production involving copper vapor and dye laser technologies, laser spectroscopy, power and tuning control.

The copper vapor laser is a gas laser excited by electric discharge. Its active medium is generated by evaporation of pieces of metallic copper placed inside the laser tube and a buffer gas (Ne or He) is added to sustain the discharge. The same electric discharge that excites the Cu atoms heats the solid copper to about 1500°C to generate Cu vapor. The laser action occurs in two different lines, with $\lambda = 5106 \text{ \AA}$ (green) and $\lambda = 5780 \text{ \AA}$ (yellow), with pulse width of 30-50 ns and peak power that easily exceeds 10 kW. The CVL development at IEAv started in 1985, with a first prototype of an externally heated copper bromide system that delivered an average power of about 100 mW at a repetition rate of 100 pps. From this system, the work evolved to self heated true CVL's, with maximum average power ranging from 5 W, from a compact air cooled system, to 40 W, for a conventional water cooled system. CVL development continues in IEAv with the study of a new technology, proposed by Jones et al.[15, 16], called HyBrID (Hydrogen Bromide In Discharge) copper laser. It

consists in adding HBr to the gas buffer; the acid reacts with pieces of copper and CuBr is formed; then, this salt is dissociated due to the impact of electrons, generating the atomic Cu vapor necessary for laser action. The authors referenced above argue that with such a technology the operation temperature can be reduced to as low as 400 °C, the efficiency raised to 3% and, contrary to the conventional CVL, the gain is concentrated at the laser tube axis, thus propitiating good quality beams. Besides the advantages above mentioned, there is still the fact that the large reduction in operation temperature allows the use of cheaper and not so noble materials for the laser heads. We built some different CVL's, with different dimensions, thermal insulation, excitation circuits, etc., in order to obtain the necessary experience in CVL design. Table 2 summarize some results obtained with CVL at IEAv [11, 17, 18, 19].

The dye laser active medium consists usually of an organic dye diluted in a solvent (ethanol, methanol, water, etc.), optically pumped by flash lamps or other laser (argon, excimer, CVL, etc.). With a set of different dyes, it is possible to obtain continuously tunable laser action from the near ultraviolet to the near infrared, achieving conversion efficiency (pumping power into laser power) above 30% in the middle of the visible range of the spectrum. Besides that, with convenient resonator designs, it is possible to obtain very narrow bandwidth laser beams. All these characteristics make this class of lasers a powerful tool for spectroscopic purposes. The dye laser can be assembled in many different configurations, depending on the required characteristics. It can operate from pulsed regime, with pulsewidth of fraction of picoseconds, to continuous wave; in the pulsed regime, it can work from single shot to high repetition pulse rate (kHz for pulsed pump until MHz for modelocking); it can emit in narrow (some MHz) or broadband (tens of GHz), and so on. The choice of the regime of operation determines the design of the pumping scheme, the dye cell and the optical resonator. In the case of AVLIS purposes, the dye laser must operate in pulsed regime, at repetition rates of about 5-20 kHz, with output peak power in the range of tens of kilowatts. Since in the range of the visible the uranium isotope shift and the ^{235}U absorption bandwidth (broadened by the hyperfine structure) lie around 5-10 GHz, the dye laser linewidth must be smaller than this limit.

All these lasers are in current use in our facilities, in experiments that range from material cutting, drilling and evaporation to pumping dye lasers for spectroscopy.

3. Although the uranium spectrum has been widely studied and several spectroscopic tables are available, most of the data have been obtained by conventional spectroscopy, and there is a huge lack of information both due the techniques used and due the great strategic significance of accurate information about laser isotope separation. Thus, we have been working at our laboratories with conventional and intermodulated laser optogalvanic spectroscopy and with laser multistep photoionization

spectroscopy [11], aiming towards the choice of the best channels leading to the stepwise photoionization.

4. Studies on light-atom interaction with emphasis on modelling, using both rate equations and the Schroedinger equation, taking into account high pulse power, short pulse duration, and the Rabi frequency have been carried out.

5. Plasma behavior and ion extraction problems also have been investigated.

6. CONCLUSIONS

Uranium isotope enrichment is universally recognized as a very expensive activity. Its development is only justified by the necessity of energy generation by nuclear reactors. The laser methods for uranium enrichment are being considered, nowadays, among the best from the economical viewpoint. They also are the best ones from the ecological point of view, since the collected tails can be extremely depleted of the fissile isotope ^{235}U .

The use of laser methods for uranium isotope separation requires, on the other hand, an amount of scientific and technological know-how which is not yet completely available. The involved problems comprise an interdisciplinary and complex knowledge that no professional alone can dominate. Studies on basic science, technology, engineering and economics are necessary, which are beyond the academic formation of any conventional technician or scientist. Nevertheless, countries which possess already the traditional technologies in advanced stages make nowadays plans of giving up [1, 20] installations that are working, in favor of investments in these new techniques. In countries where the traditional technologies have not yet been deployed in a considerable scale, it seems reasonable that new investments be directed towards technologies that can offer the possibility of being competitive in a near future in the international market.

The studies we have described before are part of a project whose aim is to demonstrate the technical viability of the laser processes for isotope separation 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 technologies. The main objectives of this work were to describe the laser processes for isotope separation and to present some useful results obtained in our laboratory. We focused our attention mainly on two actuation areas: laser development and spectroscopy. This paper presents some results obtained in these areas, such as CVL with average output power up to 40 W and the observation of 3-photon 2-frequency photoionization of uranium both in hollow cathode lamps and in a furnace.

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