Dysprosium enrichment and collection using AVLIS in Brazil: Preliminary results

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Abstract. Rare Earth elements (RE) play an important role in the high-tech industry. Displays, fine electronic devices, sensors, optoelectronics, and many other fields rely on the use of these elements to deliver the technology we depend on. The possibility of improving the capacity of all these applications is attractive and there are a few ways to achieve that, one of them is to purify the REs to get only one isotope. Given that, one of the few techniques able to separate RE isotopes is the Atomic Vapor Laser Isotope Separation (AVLIS) which is the technique used in the Brazilian facility for isotope separation via laser. In this work we are going to show the latest results on the Brazilian AVLIS applied to enrich dysprosium. As a preliminary result, it was observed an enrichment by means of a mass spectrometer coupled to the experimental setup and a thin film was collected with an enrichment factor of 1.37 ($^{163}$Dy).

1. Introduction

Dysprosium (Dy) is a Rare-Earth element that presents several high-tech applications. As a laser active medium, it has recently delivered outstanding results in mid-infrared lasers [1]. In nuclear medicine it can be used for arthritis treatment [2]. Usually, it is not used in its metallic form as it reacts with air and water, thus it is better to combine Dy in an alloy. As an example, nuclear reactors use Dy based alloys as the material for control rods, as it presents high neutron absorption without showing relevant long-term damage or shape modification [3]. Finally, super magnets (NdFeB) can benefit by the use of Dy in its alloy, as Dy-based magnets can handle higher temperatures without demagnetization [4]. Given that, this kind of magnets are the best choice for motors, generators and turbines. Figure 1 brings a chart extracted from [5] showing the concentration effect of Dy on a NdFeB magnet. The higher the Dy concentration, the higher the maximum temperature it can handle.
Figure 1. Chart of the Dy concentration effect on NdFeB magnets. $H_{cj}$ is related to magnet’s resistance to demagnetization while $B_r$ is related to magnet’s field strength. The higher these values the better [5].

All these applications can be improved using enriched material with the right selected isotope instead of the natural dysprosium. There are many ways to achieve isotope separation, the main ones are ultracentrifuges, gaseous diffusion, electromagnetic and laser techniques. Ultracentrifuges and gaseous diffusion are limited to material that are volatile at low temperatures, which is not the case of Rare-Earths (RE). Electromagnetic separation is very expensive as it consumes a lot of energy in the process. Thus, laser-based techniques are the most suitable ones for this type of elements.

Atomic Vapor Laser Isotope Separation (AVLIS) can be used to isotopically enrich theoretically any element of the periodic table, including Rare-Earths. AVLIS can deliver a very high enrichment factor which compensates the low throughput it can achieve nowadays, and this fulfils the requirements for the main RE applications (low concentrations). For that, spectroscopic data is required in order to tune correctly the photoionization lasers and excite the desired isotope to the ionized state [6,7]. Using the laser multistep excitation technique [8], the group has already observed the two first steps for dysprosium in the yellow-orange spectral range. However, Dy requires 3 photons, in the yellow-orange range, to reach autoionization states or a Rydberg state. This work shows the application of the data collected so far on a preliminary experiment to enrich dysprosium. The photoionization was achieved using a sequence of three excitation steps, tuning the wavelength of the third step, not known yet, in order to observe the effects on the photoionized isotopes, either by monitoring with a mass spectrometer coupled to the evaporation chamber or analysing deposited material on collection plates.

2. Experimental setup

The setup developed to perform the enrichment experiments is basically composed of a vacuum chamber, a photoionization laser system, a RE vaporization scheme, a coupled mass spectrometer for online-real time analyses and a collection apparatus.

A vacuum chamber was designed and produced in the Institute. It is composed of four optical windows, one transmits dye laser beam, another one is used for laser ablation (not considered in this work) and other two for imaging and general observations, of two electrical feedthroughs, one is for
the collection plates polarization and another one to apply high current into the crucible. There is also one mass spectrometer inlet operating as RGA (Residual gas analyzer).

Three Dye lasers operating in pulsed mode (pulse length, output power, repetition rate, and full width at half maximum are 10 ns, 700 mW, 5 kHz and 1.5 GHz respectively—Radiant Dyes) were used to excite the atoms. Laser emission spectrum was not optimized for RE isotope separation as its lines are broader than the Dy isotopic shifts, thus two or more nearby isotopes will be excited. The photoionization steps are the following: $\lambda_1 = 565.3$ nm; $\lambda_2 = 601.2$ nm, since previous experiments showed that as the most efficient route at the available laser range, and $\lambda_3$ was tuned over the 3rd laser available spectral range (564-571 nm). A homemade synchronization system was used to provide a temporal overlap of the three pulses. Also, it is required to deliver a mixture of three beams. For this purpose, a system composed of three optical fibers was developed. This system is composed of three fibers of 300 μm core diameter, that are bundled into a 1 mm core diameter fiber which delivers the mixed beam to the vacuum chamber through a collimation lens. The beam diameter inside the vacuum chamber is of 8 mm.

Figure 2 shows the complete experimental apparatus. This setup allows to register the photoions mass distribution in real time using a mass spectrometer (model Prisma plus Pfeiffer Vacuum) coupled to the chamber (MS). Moreover, a collection stage was mounted just above the photoionization region, where two parallel plates, with applied voltage, generate an electric field to collect the photoions. It was applied a voltage of 500 V, providing an electric field of $10^4$ V/m.

Finally, the generation of Dy vapor was obtained using a Dy metallic sample (99.9%-GoodFellow) heated in a platinum crucible by Joule effect until it reaches the vapor pressure and evaporates.

![Figure 2. Experimental apparatus to detect the isotopic signal. DL$_{1,2,3}$: dye laser (sequential excitation system), AL: ablation laser, L: lens, VC: vacuum chamber, PS: power supply, Rec: recorder, M: mirror, P: plasm plume, (dysprosium), MS: mass spectrometer and Dy: dysprosium target.](image)

The material is collected on a titanium plate and it is taken to a SIMS (Secondary Ion Mass Spectrometer - Hiden analytical) where the isotope composition is assessed. A pure dysprosium metallic sample, same as the one used for evaporation, is also measured as a standard for abundance comparison.
3. Results and discussion

As a first result it was observed the change in the photoions current inside the coupled mass spectrometer when tuning the 3rd step laser wavelength. A few third step wavelengths were observed, and the most evident isotope ratio change took place at 566.2 nm. Figure 3 shows the signal measured for two different fine-tuning conditions at this wavelength (566.2 nm – changing the second decimal place).

![Figure 3](image)

*Figure 3. Dy isotope photoions measured by the coupled mass spectrometer for on-line measurements and optimization. Each of the two curves shows measured isotope relative intensity for a different 3rd step wavelength.*

Given these observations the next step was to effectively collect any amount of enriched material. For that, the collection system was implemented into the chamber and the photoionized material collected by the negative plate. After two hours the lasers were turned off and the plate was taken to a SIMS spectrometer. By measuring the isotope composition of the plate and of a natural reference, it was possible to obtain the isotopic abundance of the collected material. Figure 4 shows these results.

![Figure 4](image)

*Figure 4. Natural Dy abundance measured in a commercial metallic sample (blue) and collected after photoionization (red).*
It was observed an enrichment of the odd isotopes $^{161}$Dy and $^{163}$Dy. The estimated enrichment factors are: $^{161}$Dy = 1.30 and $^{163}$Dy = 1.37. The amount of material collected is very small and could not be quantified. To fully understand why only odd isotopes were enriched, in the condition used in this work, further data and simulation of the hyperfine split and its position in the isotope spectrum are required.

4. Conclusions
It was demonstrated the enrichment and collection of Dy odd isotopes using the AVLIS approach. The results are preliminary and far from the high efficiencies observed in the literature for other materials, however it demonstrates good integration of the whole system and proves that the knowledge obtained so far is correct. An enrichment factor of 1.37 ($^{163}$Dy) in a unique stage was observed reinforcing the potential of the AVLIS technique. Future research is going to be performed in order to enhance this system in terms of enrichment factor and amount of material collected.

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