Measurements of the Effective Lifetime of the $^7M_7$ Level of Uranium in a Hollow Cathode Lamp by Two Different Methods: Optogalvanic and Photoionization Spectroscopy

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This paper presents the experimental investigation of the effective lifetime of the 7M7 uranium level in a hollow cathode lamp, with two different methods of population probing: using the optogalvanic signal in a DC discharge and using the photoionization signal in the afterglow of a pulsed discharge.

Este trabalho apresenta o resultado de estudos experimentais do tempo de vida do nível $^7M_7$ do urânio realizados em uma lâmpada de catodo oco, utilizando duas técnicas diferentes para a monitoração da população deste excitado: usando o efeito optogalvânico em uma lâmpada excitada por descarga contínua e usando sinais de fotoionização na pós-descarga de uma descarga pulsada.

Introduction

The Atomic Vapor Laser Isotope Separation (AVLIS) project, in development at IEAv, demands extensive studies of multi-step, multi-frequency photoionization spectroscopy in metallic U [1]. We have shown that these studies can be made in U vapor generated in pulsed hollow cathode lamps [2]. The use of hollow cathode lamps instead of vacuum furnaces simplifies the experimental apparatus and reduces the risks of working with large amounts of radioactive vapor. However, some physical parameters, important to the AVLIS process, that are different if the U vapor is produced in a hollow cathode lamp or in a vacuum furnace, mainly due to collisional processes. One of these parameters is the effective level lifetime, that affects the absorption saturation.

This paper presents the experimental investigation of the effective lifetime of the $^7M_7$ uranium level in a hollow cathode lamp, with two different methods of population probing: using the optogalvanic signal in a DC discharge and using the photoionization signal in the afterglow of a pulsed discharge. In the first case, we compare the influence of collisions of U atoms with electrons of the discharge and with atoms of the buffer gas. In the second case we eliminated the influence of collisions with electrons by measuring the effective lifetime in the afterglow. The effective lifetime obtained with optogalvanic signal in the limit of zero current compares well with the result obtained with the photoionization signal in the afterglow.

Experiments with optogalvanic signal

Two pulsed dye lasers, pumped by copper vapor lasers, are tuned to the $\lambda_1 = 5915$ Å and $\lambda_2 = 6051$ Å uranium transitions. These two wavelengths provide a two steps sequential excitation of the U atom, as indicated in Fig. 1. The two laser beams are combined and illuminate the hole of a uranium hollow cathode lamp, filled with 4.5 torr of argon as gas buffer, as shown in

Fig.2. A DC power supply sustains a continuous discharge in the hollow cathode lamp. The optogalvanic signal, due to absorption of laser radiation, is coupled into an oscilloscope and a boxcar averager.

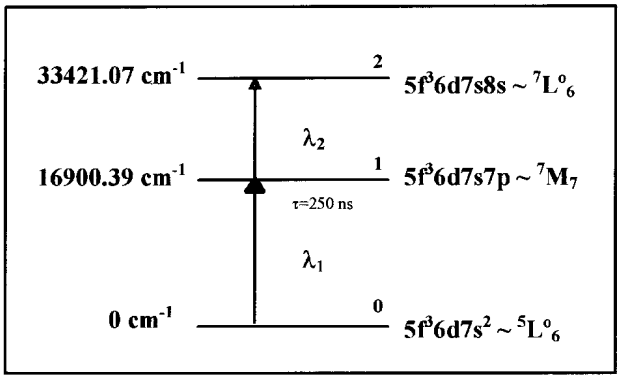

Figure 1. Diagram of the used excited levels.

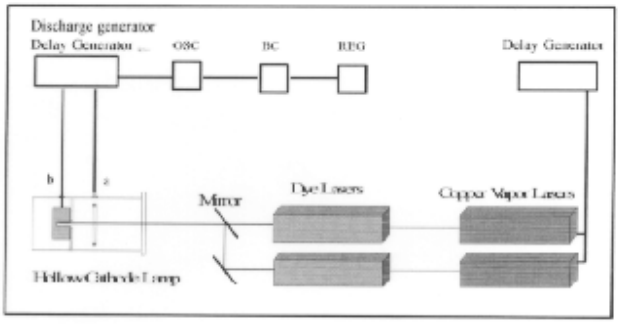

Figure 2. Experimental Setup. Oscilloscope (OSC), Boxcar (BC), Plotter (REG).

First, only laser 1 illuminates the discharge. The optogalvanic signal due to the absorption of λ_1 photons provides a background level for the electric signal. When laser 2 is released, illuminating the discharge, there is an increase in the electric signal due to the absorption of λ_2 photons. If we assume that this additional signal is proportional to the $^7M_7$ level population, it can be used as a measure of the level population in the instant the laser 2 is fired [3]. So, delaying laser 2 pulses relatively to laser 1 and measuring the peak of the electric signal (subtracting the signal background), we can trace the $^7M_7$ level population along time. Typical results for these experiments are shown in Fig.3.

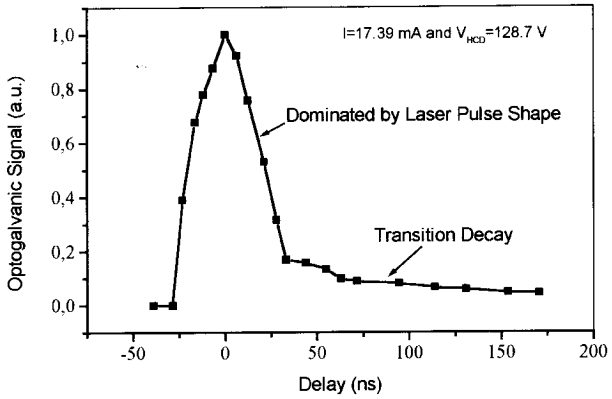

Figure 3. Amplitude of the optogalvanic signal due to the absorption of 12 photons against delay between lasers. Negative values for the time mean laser 2 was fired before laser 1; positive values mean laser 2 was fired after laser 1.

In previous works, we had shown that the curve describing the optogalvanic signal amplitude against laser delay has two characteristic regions: in the first one, the shape of the curve is determined by the laser pulse shape, while in the second one, the curve follows an exponential decay with the transition decay time [4,5,6]. Therefore, fitting the *tail of the curve* to a decaying exponential provides the effective transition lifetime for a given experimental condition.

We repeated this procedure for different values of electric discharge, obtaining this way the effective decay rate (1/teff) against the electric current density in the hollow cathode lamp, as shown in Fig.4. The decay rate augmentation with the electric current can be explained as follows.

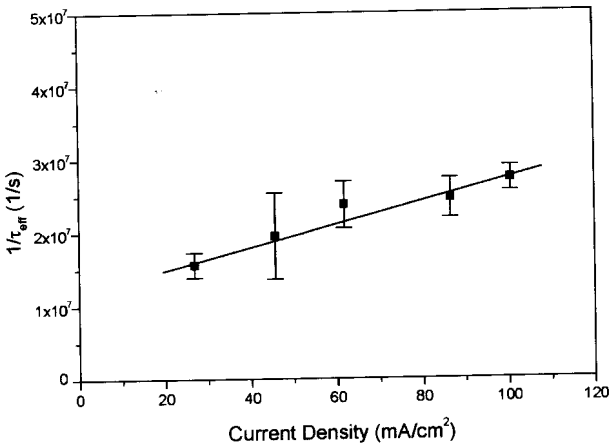

Figure 4. Effective lifetime against hollow cathode electric current density. Experiments with optogalvanic signal.

The level decay rate in a electric discharge has three main contributions, that can be written as

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau} + < \sigma\nu > Ar^n Ar + < \sigma\nu > e_e^n \qquad (1)$$

where τ is the level radiactive lifetime, $< \sigma\nu > Ar$ is the quenching rate due to uranium-argon collisions, $< \sigma\nu >_e$ is the depopulation rate due to collisions with electrons (inelastic and super-elastic collisions, ionization, excitation etc...) and n_{Ar} and n_e are the argon and electron population respectively. For a sealed hollow cathode lamp operating in the normal glow discharge, the electron density is nearly proportional to the electric current density J [3,4], and Eq. 1 can be rewritten as

$$\frac{1}{\tau_{eff}} \approx \frac{1}{\tau} + \alpha + \beta J \qquad (2)$$

where α and β are given by:

$$\alpha = \frac{16\sqrt{\pi a^2}}{\sqrt{\mu k T_0}} p$$

$$\beta = \frac{< \sigma v >_e}{e^- v_{\text{drift}}}$$

and where $a = (r_U + r_{Ar})$, r_U and r_{Ar} are the uranium and argon atom radius, respectively, μ is the reduced mass, k is the Boltzmann constant, T_0 is the absolute temperature, p is the argon pressure, $< \sigma v >_e$ is the collisional rate between uranium and argon, e^- is the electron electric charge and v_{drift} is the electron drift velocity in the plasma.

In first approximation, we consider α and β as constants. This way, the decay rate can be described as linear with J. The best fit for the experimental data in Fig.3 gives

$$1/t_{\text{eff}} = (1.2 \pm 0.1) \times 10^7 + (1.50 \pm 0.13) \times 10^5 J \ ,$$

being J in mA/cm^2 and τ_{eff} in seconds. Thus, for normal operation conditions, around 1000 mA/cm^2, the decay rate due to electron impact is in the same order of magnitude as the decay rate due spontaneous decay plus collisions with argon. Taking the limit of zero current, the effective lifetime of the ^{7}M$_7$ level of the uranium is about 83 ns. Since the radiactive lifetime of the ^{7}M$_7$ level is 250 ns [7], the quenching rate due to uranium-argon collision is about 8.0×10^6 s^{-1}.

Experiments with photoionization signal

In this case, photoionization signal was used to monitor the ^{7}M$_7$ level population instead of the optogalvanic signal. Basically the same experimental apparatus was used, except that the hollow cathode discharge was pulsed and electric signal due to photoionization observed in the afterglow of the discharge. When uranium vapor is illuminated by a laser beam tuned to 5915 Å , two different process can happen: a-) the ^{7}M$_7$ level is populated by single-photon absorption; and b-) photoionization is produced by two-photons absorption followed by single-photon absorption to the continuum [1]. The first mechanism is by far more likely to occur, with a cross section of about 10^{-13} cm^2, against 10^{-17} cm^2 for the second process. When a second laser, tuned to 6051 Å , illuminates the uranium vapor, atoms in the ^{7}M$_7$ level are ionized by absorption of single 6051 Å photon followed by absorption of a 5915 Å photon. So, in a way analog to the optogalvanic technique described before, the 5915 Å laser beam populates the ^{7}M$_7$ level while the photoionization signal due to the 6051 Å laser beam can be used to monitor the ^{7}M$_7$ level population.

Once again, the first laser determines a electric signal background and the variation of the electric signal due to the second laser is used as a measure of the ^{7}M$_7$ level population. Laser 2 pulses are delayed in respect to laser 1 and the effective lifetime is obtained by adjusting the experimental data to a decaying exponential.

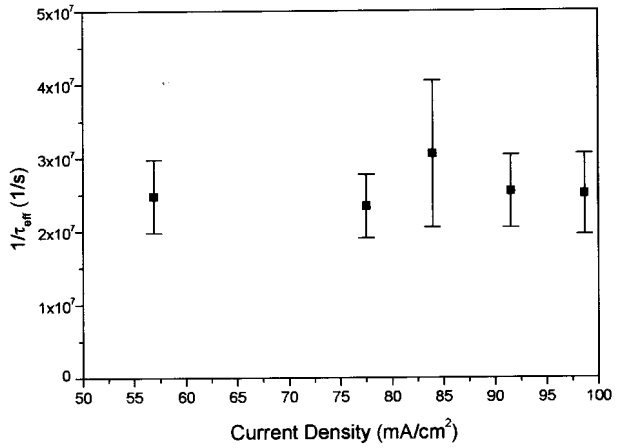

Figure 5. Effective lifetime against hollow cathode electric current density. Experiments with photoionization signal.

Fig. 5 shows effective lifetimes, obtained with this method, for different electric current densities in the pulsed discharge. It is necessary to stress out that this measured effective lifetimes must not depend directly on the electric current because the photoionization signal is measured in the afterglow, about 1 ms after the discharge is turned off. Averaging those values we have $(1/\tau_{eff}) = (2.4 \pm 0.5) \times 10^7$ s^{-1}.

Discussion

The measurements made with optogalvanic signals showed that collisions with electrons in a hollow cathode lamp discharge have the same importance as collisions with argon atoms, as far as decay rate is concerned. Both the quenching rate due to collisions with argon and due to collisions with electrons are in the order of magnitude of the spontaneous level decay rate ($1/\tau_{sp}$), making the effective lifetime of the $^7M_7$ level considerably shorter than the spontaneous lifetime in our experimental conditions.

The experiments made in the afterglow of the pulsed discharge resulted in an effective decay rate twice as big as the same parameter obtained with optogalvanic signals. This difference can be attributed to the fact that the temperature variation due to electric discharge was not considered in the analysis of the results presented in Fig.4. A detailed experimental study is still necessary to evaluate this hypothesis. Although the differences in the results, these experiments allow a good evaluation of the magnitude of the influence of collisions between uranium and argon in the effective lifetime of the $^7M_7$ level.

We concluded also that collisions must be taken into account when transposing results of spectroscopic studies made with hollow cathode lamps to AVLIS experiments performed in evaporation chambers.

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