Experimental investigation of the effective lifetime of the $^{7}\text{M}_7$ level of uranium using the optogalvanic signal in a DC discharge and photoionization signal in a pulsed discharge

José Wilson Neri, Carlos A. B. da Silveira, Nicolau A. S. Rodrigues, Marcelo G. Destro, Carlos Schwab and Rudimar Riva

Instituto de Estudos Avançados /CTA - São José dos Campos - SP

jwneri@ieav.cta.br

Abstract

This paper presents the experimental investigation of the effective lifetime of the $^{7}\text{M}_7$ 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.

Introduction

The Atomic Vapor Laser Isotope Separation (AVLIS)\(^1\) project, in development in the IEAv, demands extensive studies of multi-step, multi-frequencies photoionization spectroscopy in metallic U [1]. We have shown that these studies can be made generating the U vapor with pulsed hollow cathode lamps [2]. The use of hollow cathode lamps instead of vacuum furnaces simplifies a lot the experimental apparatus besides of reducing the risks of working with large amounts of radioactive vapor. However, there are physical parameters, important to the AVLIS, that changes 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 influences, for instance, the absorption saturation.

This paper presents the experimental investigation of the effective lifetime of the $^{7}\text{M}_7$ 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.

Results and Discussions

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.

Figure 1: Diagram of the used excited levels.

\(^1\) Called Processo Atômico de Separação Isotópica a Laser (PASIL) in IEAv.
The two laser beams are combined and illuminate the hole of a uranium hollow cathode lamp as shown in Fig. 2. Three Uranium Hollow Cathode Lamp (HCL) were used with natural uranium cathode and filled with Argon gas as buffer to the discharge: HCL-1 (p=2.8 mbar, \( f = 0.30 \) cm, \( l = 0.9 \) cm), HCL-2 (p=3.1 mbar, \( f = 0.47 \) cm, \( l = 1.7 \) cm), HCL-3 (p=4.5 mbar, \( f = 0.30 \) cm, \( l = 1.0 \) cm). A DC power supply sustains a continuous discharge in the hollow cathode lamp and the optogalvanic signal is measured.

![Electric Circuit Diagram](image)

**Figure 2:** Experimental Setup. Boxcar (BC); Copper Vapor Laser (CVL); Dye Laser (DL); Oscilloscope (OSC); Generator Delay (G), Plotter (REG); Mirror (M)

The optogalvanic signal due to the absorption of \( \lambda_1 \) photons provides a background level. The signal fluctuation due to the absorption of \( \lambda_2 \) photons is used to monitor the population of the \(^7\text{M}_2\) level. So, delaying the \( \lambda_2 \) laser pulses in relation to the \( \lambda_1 \), we can follow the time behavior of the \(^7\text{M}_2\) level population. We had previously shown that the effective lifetime of this level can be obtained by adjusting the experimental data of the tail of the “optogalvanic signal against delay time curve” to a decaying exponential [3,4,5].

Figure 3 shows the \( 1/\tau_{\text{eff}} \) in function of the \( \lambda J/p \) parameter. Considering the \( \lambda J/p \) as an invariant parameter for the hollow cathode lamp [6].The solid line is a linear fitting.

![Data Plot](image)

**Figure 3:** Uranium \(^7\text{M}_2\) level effective lifetime versus hollow cathode discharge \( \lambda J/p \). Experiments with optogalvanic signal

The decay rate augmentation with the electric current can be explained as follows. The level decay rate in a electric discharge has three main contributions that can be written as:

\[
\frac{1}{\tau_{\text{eff}}} = \frac{1}{\tau_{sp}} + \langle \sigma v \rangle_{Ar} n_{Ar} + \langle \sigma v \rangle_{e} n_{e}
\]  

(1)

where \( \tau_{sp} \) is the level radiative lifetime, \( \langle \sigma v \rangle_{Ar} \) is the quenching rate due to uranium- argon collisions, \( \langle \sigma v \rangle_{e} \) 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 proportional to the electric current density \( J \) [7,3], and Eq. 1 can be rewritten as
\[
\frac{1}{\tau_{\text{eff}}} = \frac{1}{\tau_{\text{sp}}} + \alpha + \beta J
\]  \hspace{1cm} (2)

where \( J \) is the current density and:

\[
\alpha = \langle \nu \rangle_{\text{Ar}} n_{\text{Ar}} A \frac{16\sqrt{\pi} a^2 P}{\sqrt{nkT_0}} \quad \quad \beta = \langle \nu \rangle_{e} n_{e} \frac{e \nu_{\text{drift}}}{eV_{\text{drift}}}
\]

or,

\[
\alpha = A - \frac{1}{\tau_{sp}} \quad <\sigma v>_{\text{Ar}} n_{\text{Ar}} \\
\beta = \frac{\lambda}{P} = <\sigma v >_e \frac{n_e}{J}
\]

where \( A \) and \( B \) are the linear and angular coefficients of the fitted straight line and \( v_{\text{drift}} \) is the drift velocity of electrons in argon. In experimental conditions similar to ours, its obtained \( v_{\text{drift}} < 0.2 \times 10^6 \) cm/s [8].

The average energy of the electrons in a uranium hollow cathode discharge is in general low. For our experimental conditions (dump currents from 50 mA to 200 mA, \( T_e \approx 4000 \) K), \( kT_e \approx 0.3 \) eV [9].

In first approximation, we consider \( \alpha \) and \( \beta \) 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:

\[
\frac{1}{\tau_{\text{eff}}} = (1.89 \times 10^7 \pm 1.27 \times 10^6) \pm (2.20 \times 10^5 \pm 3.97 \times 10^4) J
\]

being \( J \) in mA/cm\(^2\) and \( \tau_{\text{eff}} \) in seconds. Thus, for normal operation conditions, around 50 mA/cm\(^2\), \( \lambda J/p = 15 \), the decay rate due to electron impact is in the same order of magnitude as the decay rate due to collision with argon.

Taking the limit of zero current, the effective lifetime of the \( ^7\text{M}_7 \) level of the uranium is about 55 ns. Since the radiative lifetime of the \( ^7\text{M}_7 \) level is 250 ns [10], the quenching rate due to uranium-argon collision is about \( 1.5 \times 10^7 \) s\(^{-1}\).

**Experiments with photoionization signal**

In this case the same experimental apparatus and procedure were used, except that the hollow cathode discharge was pulsed and electric signals due to photoionization were observed in the afterglow of the discharge, instead of optogalvanic signals. The measured electric signals came up because the sequential absorption indicated in Fig. 1 is followed by absorption of either \( \lambda_1 \) or \( \lambda_2 \), leading to photoionization. We delayed the laser 2 in relation to laser 1 and took the photoionization signal to monitor the \( ^7\text{M}_7 \) level population. The effective lifetime is obtained adjusting the experimental data to a decaying exponential.

Fig. 4 shows effective lifetimes, obtained using this method, for different electric current density, \( \lambda J/p \), on the pulsed discharge for the HCL-3.

![Figure 4: Effective lifetime versus hollow cathode discharge \( \lambda J/p \). Experiments with photoionization signal.](image-url)
It is necessary to stress that this measured effective lifetime 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:

\[
\frac{1}{\tau_{\text{eff}}^1} = (2.4 \pm 0.5) \times 10^7 \text{ s}^{-1}.
\]

The measured effective lifetime \(1/\tau_{\text{eff}}^0\) or \(\tau_{\text{eff}} = 40 \text{ ns}\) lower than the spontaneous lifetime \(t_{\text{sp}} = 250 \text{ ns}\) can be understood taking in consideration that the buffer gas is always present in the discharge. However, using a pulsed discharge hollow cathode lamp, we can increase the density discharge current, increasing, hence, the uranium vapor density at ground state without increasing the deleterious discharge effects.

Conclusions

The measurements made with optogalvanic signals showed that collisions with electrons in a hollow cathode lamp discharge has about the same importance as collisions with argon, 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 \(1/\tau_{\text{sp}}\), making the effective lifetime of the \(^7\text{M}_7\) level considerably shorter than the spontaneous lifetime.

The experiments made in the afterglow of the pulsed discharge resulted in a effective decay rate that is greater then the same parameter obtained with optogalvanic signals. This difference can be attributed to the fact that the temperature fluctuation was not considered in the analysis of the results presented in Fig. 3. A detailed experimental study is still necessary to evaluate this hypothesis. Anyway, these results allows the evaluation of the magnitude of the influence of collisions between uranium and argon in the effective lifetime of the \(^7\text{M}_7\) level.

References


