EXPERIMENTAL INVESTIGATION OF THE U-AR AND U-ELECTRONS COLLISIONS RATES USING A URANIUM-ARGON HOLLOW CATHODE LAMP

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Abstract

This paper presents the experimental investigation of the U-Ar and U-electrons collisions rates using the effective lifetime measurements of the uranium $^7M_7$ level obtained from optogalvanic signal in a hollow cathode lamp (HCL) working with a DC discharge.

Introduction

The resonant absorption of radiation by atoms or molecules shows in a self-sustained discharge changes in its electrical properties. These changes are observed as an increase or decrease in the conductivity of the discharge and is known as the optogalvanic effect [1]. The hollow cathode lamp is a very versatile, compact, simple and reliable tool to provide metal vapor for spectroscopic purposes. Collisions of discharge ions with cathode walls can provide a dense vapor of the cathode material. The cathode material absorption spectrum can be obtained by illuminating the discharge with the beam of a tunable laser and monitoring the optogalvanic signal versus laser wavelength.

This paper presents the experimental investigation of the U-Ar and U-electrons collisions rates using the effective lifetime measurements of the uranium $^7M_7$ level obtained from optogalvanic signal in a hollow cathode lamp (HCL) working with a DC discharge. The experimental results are in good agreement with theoretical values obtained using models found in literature.

Theory

The principles of two photon optogalvanic spectroscopy to measure the first excited level can be easily understood using the scheme shown in Figure 1 where we show the uranium levels used in this work. We measure the total optogalvanic signal as function of the delay between the two lasers. In principle this measurement can give the lifetime value of the first level excited by laser one [2].

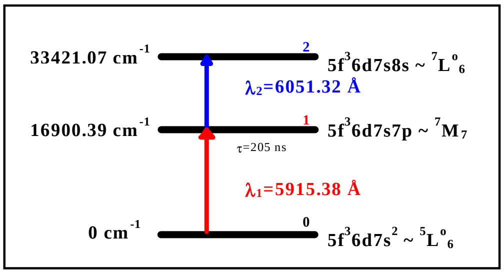

Figure1: *Excited levels diagram used in this work.*

The effective lifetime can be written as:

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau_{sp}} + \langle \sigma v \rangle_{Ar} n_{Ar} + \langle \sigma v \rangle_e n_e \qquad (1)$$

where τ_{sp} is the radiative lifetime; $\langle \sigma v \rangle_{Ar}$ is the quenching rate coefficient due the Argon - Uranium collisions; $\langle \sigma v \rangle_e$ is the electron rate coefficient including the superelastic, inelastic and ionization process due the electron - Uranium collisions; n_{Ar} and n_e are the argon and electron density, respectively. The hollow cathode lamp works in the region known as a normal glow discharge. In this condition the voltage is relatively constant for currents between 1 and 100 mA [1]. Thus, we assume that the drift velocity is approximately current independent. Besides, we assume that $\langle \sigma v \rangle_e$ is current independent, too. In this way, we can rewrite the Eq (1) as:

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

where ***J*** is the current density and [2]:

$$\alpha = \langle \sigma v \rangle_{Ar} n_{Ar} = \frac{16\sqrt{\pi}\, a^2}{\sqrt{\mu k T_o}} P \qquad \beta = \langle \sigma v \rangle_e n_e = \frac{\langle \sigma v \rangle_e}{e v_{\mathrm{drift}}}$$

Experimental Setup

The experimental setup is shown in Figure 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, ϕ=0.30 cm, ℓ=0.9 cm), HCL-2 (p=3.1 mbar, ϕ=0.47 cm, ℓ=1.7 cm), HCL-3 (p=4.5 mbar, ϕ=0.30cm, ℓ=1.0 cm).

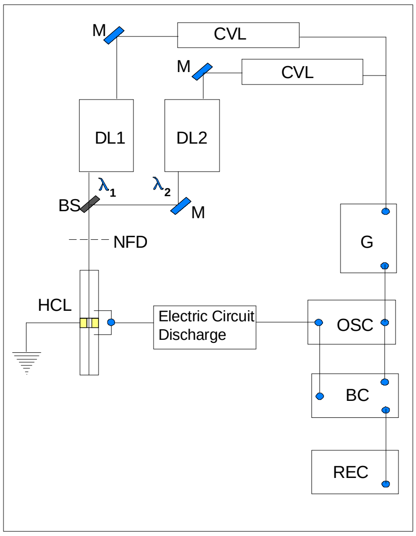

Figure 2: *Experimental setup. Boxcar (BC); Copper Vapor Laser (CVL); Dye Laser (DL); Osciloscope (OSC); Gerator Delay (G), Plotter (REG); Mirror (M)*

Results and Discussions

It has been experimentally observed that when some discharge parameters were plotted as a function of the applied voltage the data points fall onto the same curve. Therefore, this curve is denominated the discharge characteristic.

Using similarity relations, it was possible to shown that, for the same gas and cathode material, these parameters depend only on the discharge voltage, i.e., they are invariant parameters. Thus, the graphic representation of the invariant parameters allows a comparison between similar discharges. By similar discharges we mean discharges which have the same geometry, but different dimensions and are operated at different pressures.

A plot of the discharge voltage as function of $\ell J/p$, for different Ar-U hollow cathode lamp, is shown in figure 3 [3]. This shows that $\ell J/p$ is an invariant parameter for the hollow cathode lamp.

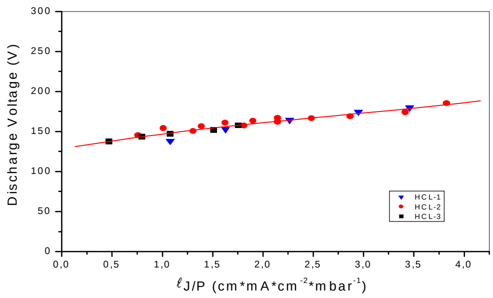

Figure 3*: Characteristic (V $\times$ $\ell J/p$) plot for three Ar-U hollow cathode discharges.*

Considering the $\ell J/p$ applied to the HCL as parameter, had been gotten some data sets for optogalvanic signal in function of the delay between the two lasers[4]. Figure 4 shows the $1/\tau_{eff}$ in function of the $\ell J/p$ parameter. The solid line is a linear fitting.

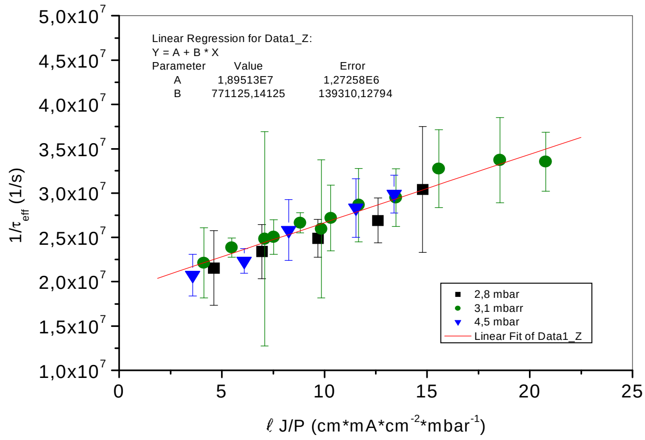

Figure 4: *Uranium $^7M_7$ level effective lifetime versus hollow cathode discharge $\ell J/p$*

Table 1 shows the experimental results obtained from the data in figure 4, where:

$$\alpha = \mathrm{A} - \frac{1}{\tau_{sp}} = \langle\sigma v\rangle_{Ar}\, n_A$$

$$\beta = \mathrm{B}.\frac{\ell}{P} = \langle\sigma v\rangle_e \frac{n_e}{J}$$

$$\langle\sigma v\rangle_e = \beta.e.v_{drift}$$

being A the B are the linear and angular coefficients of the fitted straight line and v_{drift} is the drift velocity of electrons in argon.

In experimental condicions similar to ours, its obtened $v_{drift}<0{,}2\mathrm{x}10^6$ cm/s [5].

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 **[6]**.

Table 1: *Experimental results.*

$\alpha = \mathrm{A} - \frac{1}{\tau_{sp}}$	β $\left(P/\ell \cong 3{,}5\right)$	$\langle\sigma v\rangle_{Ar} n_{Ar}$ (1/s)	$\langle\sigma v\rangle_e n_e$ (1/s) (J=50 mA/cm^2)	$\langle\sigma v\rangle_e$ (1/cm^{3}s) ($v_{drift} = 0{,}15\mathrm{x}10^6$ cm/s)
$1{,}89\mathrm{x}10^7 \pm 1{,}27\mathrm{x}10^6$	$2{,}20\mathrm{x}10^5 \pm 3{,}97\mathrm{x}10^4$	$1{,}49\mathrm{x}10^7 \pm 1{,}27\mathrm{x}10^6$	$1{,}10\mathrm{x}10^7 \pm 1{,}98\mathrm{x}10^6$	$4{,}61\mathrm{x}10^{-9} \pm 8{,}40\mathrm{x}10^{-10}$

The term $\langle\sigma \mathrm{v}\rangle_e$ in Eq (1) is due to the rates of excitation and desexcitation of the first excited level due to collisions with electrons. To estimate the U excitation and desexcitation taxes, we can consider [4]:

$$\langle\sigma \mathrm{v}\rangle_e n_e N_1 = \sum_{\substack{i=0\\ i\neq 1}}^{\infty} \langle\sigma_{1\to i}\mathrm{v}\rangle_e n_e N_1 - \sum_{\substack{j=0\\ j\neq 1}}^{\infty}\left[\sum_{\substack{k=0\\ k\neq j}}^{\infty}\langle\sigma_{j\to k}\mathrm{v}\rangle\right] n_e N_j$$

where the first term of the right side of this equation must be the excitation and desexcitation of U atoms presents in the first excited level, for all the other levels of energy, provoked by collisions with electrons in the discharge. The second term must be due to and desexcitacion of atoms of Uranium, that come from other states of energy to level 1. Making some considerations we have [4]:

$$\langle \sigma v_e \rangle = \Phi f_{ij} \left(\frac{E_H}{E_{ij}} \right)^{3/2} \left(\frac{E_{ij}}{k_B T_e} \right)^{1/2} \Psi(u_{ij}) \quad (3)$$

with:

$$\Phi = 8\pi a_0^2 \sqrt{\frac{2E_H}{\pi m_e}} = 8{,}6866x10^{-8} \text{ (cm}^3\text{/s)}$$

$$\Psi(u_{ij}) = \frac{e^{-u_{ij}}}{1+u_{ij}} \left\{ \frac{1}{20+u_{ij}} + \ln\left[1{,}25\left(1+\frac{1}{u_{ij}}\right)\right]\right\}$$

$$u_{ij} = \frac{E_{ij}}{k_B T_e}$$

$$E_{ij} = E_j - E_i$$

Figure 5 shows the total collision rates ($<\sigma v>_e$) between electrons and Uranium atoms, calculated theoretically (Eq. (3)), in function of electrons energy, for a broad range energy. The dashed region represents the typical range for average energy of the electrons in a uranium hollow cathode discharge [4], for the diverse on levels of energy to state $^7M_7$ of the uranium.

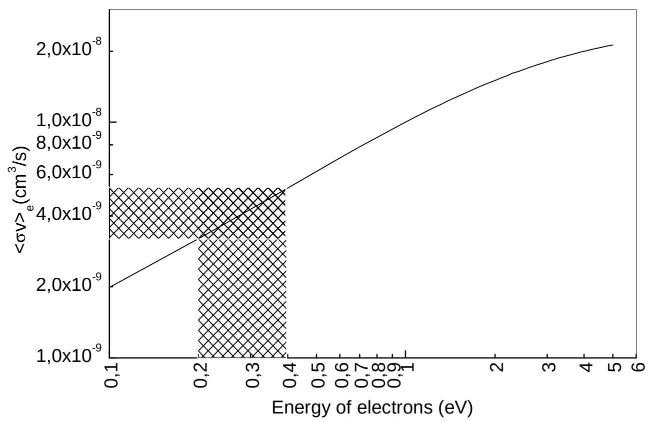

Figure 2: *Total collision rate calculated theoretical of electrons with atoms of uranium in state $^7M_7$, in function of the energy of electrons presents in the discharge.*

Conclusions

We can estimate the Uranium-Argon colisional and the Uranium-electrons colisional rates using two photon optogalvanic spectroscopy techniques.

We verify that the frequencies of collisions with argon or electrons are of the same order of magnitude for the current densities used in this work (≈ 50 mA/cm^2). The value of the rates of collision with electrons $<\sigma v>_e$ is comparable to the theoretical value, for the interval of energy of electrons that keep the discharge of hollow cathode of Uranium (≈ 0,3 eV).

References

[1] Barbieri, N. Beverini, and A. Sasso, “Optogalvanic Spectroscopy”, Reviews of Moderns Physics, 12-3, 603, 1990.

[2] N.A.S. Rodrigues, J.W. Neri, C.A.B. Silveira, M.G. Destro, C. Schwab, R. Riva, “Measurements of the effective lifetime of the $^7M_7$ level of uranium in a hollow cathode lamp”, Symposium on Lasers and Their Applications, Campinas, Dez. 1997.

[3] Scheid, V. H. B.; Neri, J. W.; Rodrigues, N. A. S.; Riva, R.; Schwab, C.; Destro, M. G.; Silveira, C. A. B. “Characteristics of hollow cathode discarges: the invariant parameter” In. Encontro Brasileiro De Física De Plasmas, 1998, Águas de Lindóia, SP Proceedings of the 5º Encontro Brasileiro de Física de Plasmas, São Paulo - SP Sociedade Brasileria de Físca, 1998, p. 61-64.

[4] José W. Neri, “Espectroscopia Optogalvânica e de Fotoionização a Múltiplos Passos em Lâmpada de Catodo Oco de Urânio Metálico”, Tese de Doutorado, ITA, (junho 1998)

[5] C.B. Sanborn, “Basic Data of Plasma Physics”, The M.I.T. Press (1959).

[6] R.A. Keller, B.E. Warner, E.F. Zalewski, P. Dyer, R. Engleman Jr., and B.A. Palmer, J. de Physique 44, C7-23, 1983.