Uranium $^7M_7$ level lifetime measurements by two-photons optogalvanic spectroscopy

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Abstract

Two-photon optogalvanic spectroscopy technique was used to measure the lifetime of the first excited state of uranium atoms contained in a hollow cathode discharge. This was accomplished from measurements of the optogalvanic signal magnitude as a function of the time delay between two laser pulses. Preliminary results were obtained using this technique to the uranium $^7M_7$ excited level.

Introduction

The resonant absorption of radiation by atoms or molecules present in a self-sustained discharge changes its electrical properties. This change is observed as an increase or decrease in the conductivity of the discharge and is known as *the optogalvanic effect* [1]. This effect has been shown to be a powerful and inexpensive technique for investigation of atomic and molecular species, and is particularly useful in the spectroscopy of refractory elements, like uranium. Many extensive theoretical and experimental works have been published in the last years showing the results that can be obtained from optogalvanic spectroscopy [1-9]. This technique may give useful information about high-lying levels assignment and for measurements of isotope shifts, fine and hyperfine structures, etc. However great care must be taken in the utilization of the data for the determination of line shape and linewidth [1].

The main purposes of the present work are to explain the measurements of the uranium first excited level lifetime using two-photon

optogalvanic spectroscopy techniques and to investigate how important are the collisional effects in hollow cathode discharge kinetics. Although there are many other techniques [10 -12] that can be used to obtain the lifetime of atomic energy levels with more accuracy than optogalvanic spectroscopy, this later technique becomes attractive due to its simplicity and to the low cost of the experimental apparatus, compared to the former referred techniques.

In section 2 the principles of the two-photon optogalvanic spectroscopy and the theoretical model that is used are presented. In section 3 the experimental setup is described and in section 4 the obtained results are discussed. Finally the paper is concluded with some considerations about the advantages and limitations of this technique.

Theory

The principles of two-photon optogalvanic spectroscopy applied to level lifetime measurements can be easily understood using the scheme shown in Figure 1, for the uranium levels considered in this work. The uranium atoms are supplied by cathodic sputtering in an uranium hollow-cathode discharge lamp filled with argon gas as buffer. In order to measure the $^7M_7$ U lifetime and to observe the effects of electrons and atoms collisions on it, a first laser, tuned to the wavelength 5915.38 Å, is used to excite uranium atoms lying in the ground state. In addition, a second laser, tuned to 6051.32 Å, is used to stimulate the transition to the higher energy level $^7L^\circ_6$, according to Figure 1. These two wavelengths are kept fixed during the experiments. With such conditions the total optogalvanic signal is measured as a function of the time delay between the two laser pulses. As the magnitude of the signal is proportional to the atomic density of this level, these measurements give the effective lifetime value of the $^7M_7$ level.

The rate equations for this three levels scheme are given in Eqs. 1 where N_i is the i^{th} level density; $W_{i,j}$ is the electron excitation rate from i to j levels; I_i is the i^{th} laser intensity; I_{sij} is the saturation intensity of the transition from i to j levels and τ_{ij} is the transition effective lifetime. Considering the first of Eqs. 1, the first two terms are due to the electron excitation rate, the next two terms are the laser stimulated absorption and emission rates and the last two terms are due to the spontaneous decay.

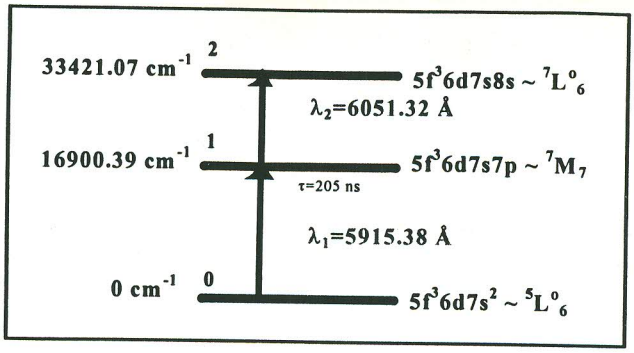

Figure 1: Diagram of the considered levels of uranium.

$$\frac{dN_1}{dt} = W_{0,1}N_0 - W_{1,2}N_1 - \frac{I_1}{I_{s0,1}}\frac{(N_1 - N_0)}{\tau_{1,0}} + \frac{I_2}{I_{s1,2}}\frac{(N_2 - N_1)}{\tau_{2,1}} - \frac{N_1}{\tau_{1,0}} + \frac{N_2}{\tau_{2,1}}$$

$$\frac{dN_2}{dt} = W_{0,2}N_0 + W_{1,2}N_1 - \frac{I_2}{I_{s1,2}}\frac{(N_2 - N_1)}{\tau_{2,1}} - \frac{N_2}{\tau_{2,1}} - \frac{N_2}{\tau_{2,0}} \quad (1)$$

$$N_{total} = N_0 + N_1 + N_2 = const.$$

Figure 2 was obtained by numerically solving Eqs. 1 using the 4th order Runge-Kutta method, considering typical values of our experimental parameters. The solution of Eqs.1 shows that when the lasers pulsewidths are smaller than the effective lifetime τ_{eff}, this parameter can be obtained, as expected, by adjusting the decay of the total optogalvanic signal to an exponential function. For pulsewidths larger than τ_{eff} the fitting should be made with the tail data points. This is the case of the present work, as will be shown in section 3.

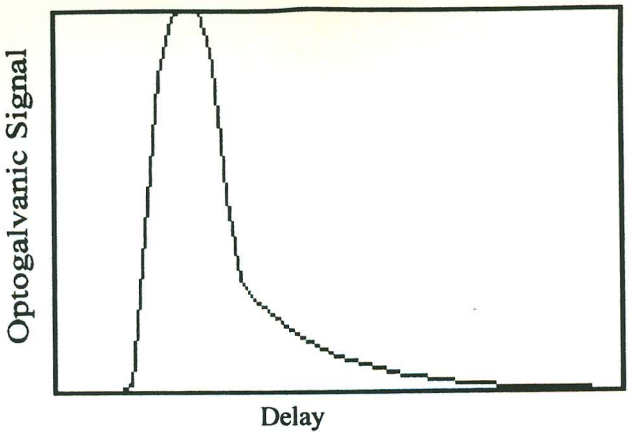

Figure 2: Result obtained solving Eqs. 1 using the 4th order Runge-Kutta method, considering typical values of our experimental parameters.

The effective lifetime can be written as:

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau} + \langle \sigma v \rangle_{Ar} n_{Ar} + \langle \sigma v \rangle_{e} n_{e} \qquad (2)$$

where τ is the radiative lifetime; $\langle \sigma v \rangle_{Ar}$ is the quenching rate coefficient due to argon - uranium collisions; $\langle \sigma v \rangle_{e}$ is the electron rate coefficient including the superelastic, inelastic and ionization processes due to the electron - uranium collisions; n_{Ar} and n_e are the argon and the electron densities, respectively. As the uranium - argon collisional term can be described by two colliding rigid spheres model, the second term in Eq. (2) is given by [13]:

$$\frac{1}{\tau_{c_{Ar}}} = \langle \sigma v \rangle_{Ar} n_{Ar} = \frac{16\sqrt{\pi}\, a^2}{\sqrt{\mu\, k\, T}}\, p \qquad (3)$$

where p is the buffer gas pressure; μ is the reduced mass; k is the Boltzmann constant; T is the system temperature and a is the collisional radius which is taken as the distance between the centers of the atoms during the collision time [14].

The electron density n_e can be expressed as a function of the current density J and the drift velocity v_{drift} as [13]:

$$n_e = \frac{J}{e\, v_{drift}} \qquad (4)$$

Thus the third term in Eq. (2) can be given by:

$$\frac{1}{\tau_{c_{ne}}} = \langle \sigma v \rangle_e \frac{J}{e\, v_{drift}} = \frac{\langle \sigma v \rangle_e}{A e\, v_{drift}} I \qquad (5)$$

where A is the cathode area. The hollow cathode lamp was operated in a regime known as a normal glow discharge. In this condition the voltage is relatively constant for currents between 1 and 100 mA [1]. Thus, it is assumed that the drift velocity and the term $\langle \sigma v \rangle_e$ are approximately current independent, so that Eq. (2) can be written as:

$$\frac{1}{\tau_{eff}} \approx \frac{1}{\tau} + \alpha + \beta I \qquad (6)$$

where:

$$= \frac{16\sqrt{\pi}\, a^2}{\sqrt{\mu k T_o}} p \quad \text{and} \quad \beta = \frac{\langle \sigma v \rangle_e}{A\, e\, v_{drift}} . \qquad (7)$$

Eq. (6) shows that the effective lifetime is described by a linear function of the current in the lamp. If the effective lifetime is measured as a function of the hollow cathode discharge current, the collisional parameters α and β can be obtained by adjusting the measured data according to Eq.6, once the radiative lifetime is known.

Experimental Setup

The experimental setup is shown in Figure 3. Two copper vapor lasers (CVL) were used to pump two Molectron DL14P dye lasers (DL1 and DL2). The dye lasers could deliver average powers of about 100 mW at 5 kHz pulse rate frequency, with 25 and 43 ns pulsewidth and with 700 MHz linewidth. A generator (G) was used to synchronize and to provide a time delay between the trigger of the two lasers. It also supplied the signal used to trigger a Digitizing Oscilloscope (OSC) (Tektronix mod.TDS-744A) and a Boxcar Averager (BC). The OGS was recorded by a HP 7046B-XY plotter (REC) from a signal f(t) supplied by the Boxcar. The uranium lamp (HCD), constructed at IPEN-CNEN/SP, contained a cathode of natural uranium that was a cylindrical piece with a 9 mm long and 3 mm diameter hole on its axis and was filled with 2.1 Torr argon buffer gas. The dc current and voltage were supplied by a stabilized

power supply (S). A half-meter Spex Monochromator (M) was used to obtain rough calibration spectra at real time.

Results

Figure 4 shows a typical two photons optogalvanic signal amplitude as a function of the time delay between the two dye laser pulses, for a discharge of 17 mA and 128 V in the hollow cathode lamp. The curve obtained shows two distinct regions, one dominated by the laser pulse shape and the other, located at the tail of the signal, dominated by the first excited level transition decay. This experimental signal shape has a good functional agreement with the theoretical behavior obtained from the rate equations solution described in section 2.

Therefore, as the laser pulsewidths have the same order of magnitude of the effective lifetime, only the tail points of the curve were used to fit the exponential decay function. Figure 5 shows the experimental data and the exponential fitting to five different currents.

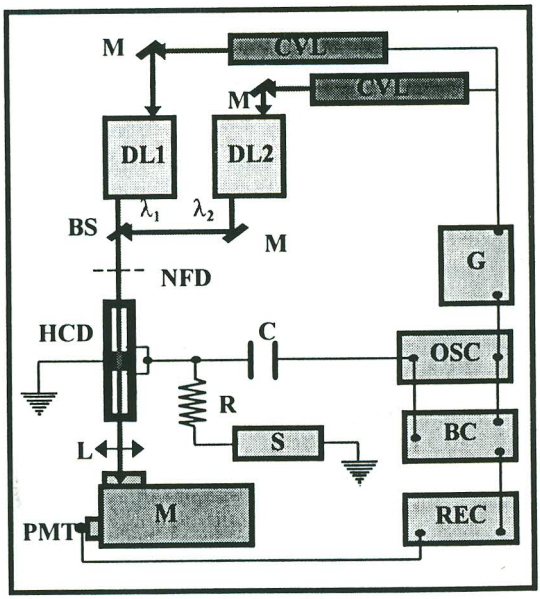

Figure 3: Experimental setup.

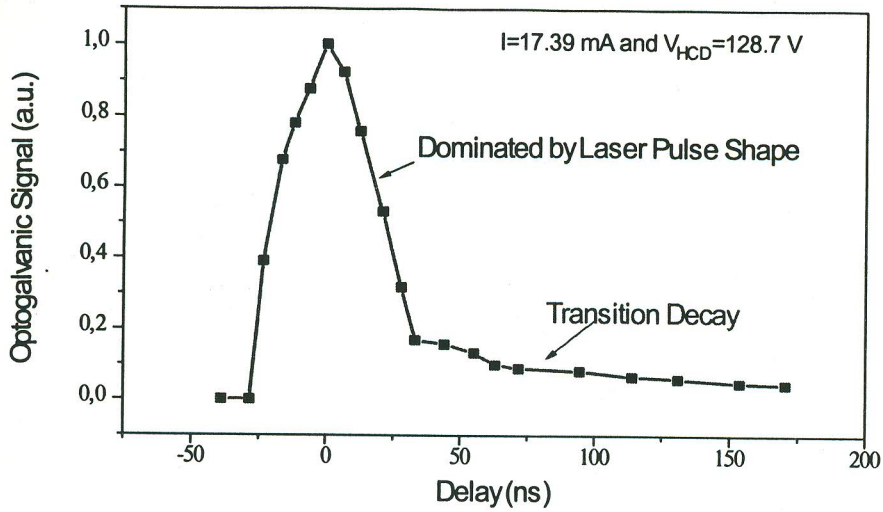

Figure 4: Two-photon optogalvanic signal versus delay between two laser pulses.

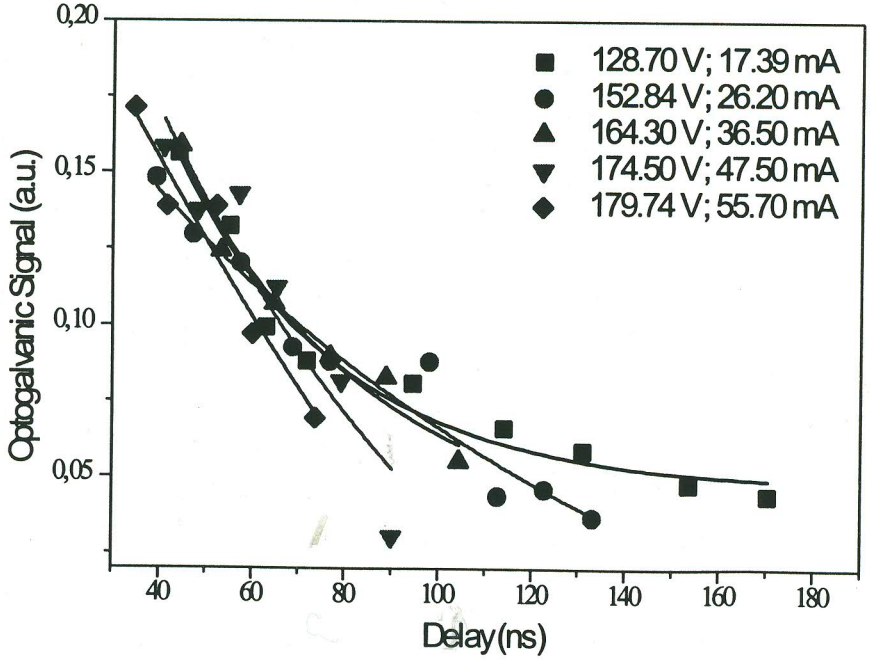

Figure 5: Exponential decay of the $^{7}M_{7}$ level.

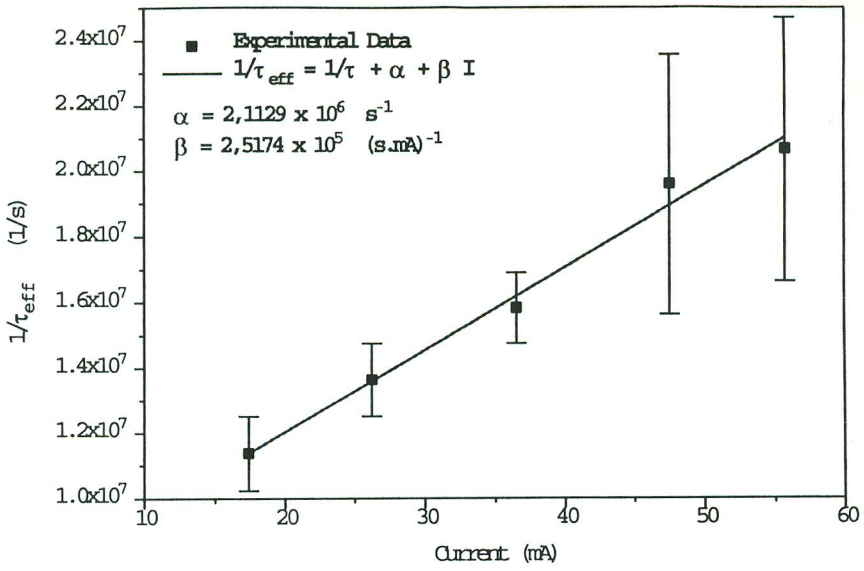

Figure 6: Uranium $^7M_7$ level effective lifetime versus hollow cathode discharge current.

In Figure 6 the effective lifetimes obtained from Figure 5 were plotted as a function of the discharge current. One sees a good agreement between the experimental data and the theoretical fitting. This agreement points out that the assumptions made in section 2 are valid for our experimental condition. Using the method of least squares to fit the experimental data we obtained the U-Ar collisional rate ($\alpha = 2.11 \times 10^6 s^{-1}$) and the electron collision coefficient ($\beta = 2.52 \times 10^5 s^{-1}.mA^{-1}$). From the α value it was possible to calculate the impact parameter of U-Ar as being equal a 0.6 Å. The order of magnitude of this value agrees with atomic radius for argon and uranium. From the β value the uranium-electron collisional rate, $\langle \sigma v \rangle_e$, was obtained as being approximately equal to 7.5×10^{-7} $cm^3 s^{-1}$ for I = 30 mA and $n_e \sim 10^{13}$ cm^{-3} [15]. This result is approximately two orders of magnitude greater than the theoretical value, obtained using a Maxwellian approximation to the electron distribution energy [16 , 17]. This disagreement is probably due to the fact that we do not consider all the ways the first excited levels can be de-excited by electron impact.

Conclusions

It was possible to estimate the uranium-argon and the uranium-electrons collisional rates using two-photon the optogalvanic spectroscopy technique. It can also be seen, using two-photon optogalvanic spectroscopy, that the electron collisional effect is the predominant factor that must be considered in lifetime measurements. Besides, the theoretical assumptions made for the collisional terms gave a good agreement with the experimental data, within the current range used in this work. Experiments varying the buffer gas pressure are necessary if one intends to measure the spontaneous lifetimes using this technique. Although there are several other techniques that can be used to measure the excited level lifetime with more accuracy than two-photon optogalvanic spectroscopy, the later can be used to give a good understanding of the collisional effects in a hollow cathode discharge and also to give an approximate result to the excited level lifetime with a relatively simple and cheap experimental setup.

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