IL NUOVO CIMENTO Vol. 14 D, N. 6 Giugno 1992

Determination of the Saturation Parameter of Electronic Transition in a Uranium-Neon Hollow-Cathode Discharge by Optogalvanic Spectroscopy.

A. Mirage (1), D. Pereira (2), F. C. Cruz (2) and A. Scalabrin (2)

(1) *Comissão Nacional de Energia Nuclear, Instituto de Pesquisas Energéticas e Nucleares C. Postal 11049 (Pinheiros) - 05499 São Paulo, SP, Brazil*
(2) *Instituto de Física Gleb Wataghin, UNICAMP - 13081 Campinas, SP, Brazil*

(ricevuto il 7 Gennaio 1992; approvato il 6 Febbraio 1992)

Summary. — The optogalvanic signal (OGS) induced in a uranium-neon hollow-cathode discharge was measured as a function of the laser power density for the $0 \rightarrow 16900\ \text{cm}^{-1}$ (591.5 nm) uranium transition. Theoretical relations derived by solving a two-level system rate equations showed the OGS dependence on the laser photon flux, for a modulated c.w. light and for stimulated transitions starting from the ground state. A fitting of the theoretical relations to the experimental measurements allowed the determination of the $\sigma_0 \tau$ product, that is, the saturation parameter of the transition. The results showed good agreement between the $\sigma_0 \tau$ values obtained by the optogalvanic and the usual optical absorption processes.

PACS 32.90 – Other topics in atomic spectra and interactions of atoms with photons.
PACS 32.80 – Photon interactions with atoms.
PACS 35.80 – Atomic and molecular measurement and techniques.

1. – Introduction.

Impedance changes can be induced in a gas discharge tube by laser radiation resonant with electronic transitions of species present in the plasma [1]. The two major mechanisms that contribute to this effect (optogalvanic effect) have been pointed out by Keller and Zalewski [2], Drèze *et al.* [3] and Kopeika [4]. The first two authors consider the increase in the plasma temperature through electron collisions with atoms excited by laser radiation as the main mechanism that causes the impedance change. Kopeika, on the other hand, considers the possibility of atomic excitation followed by ionization due to collisions with other species, thus allowing the possibility of selective ionization of chosen atoms.

It is generally assumed that both mechanisms above do occur simultaneously and that the contribution of each mechanism depends on the levels involved in the transition and on the experimental conditions. Since it is very difficult to establish a

PTC aut°

complete relationship of the optogalvanic signal to the several discharge parameters, some simplified mathematical models have been developed that could correctly describe the observed results for some specific experimental conditions [5-9]. A great deal of theoretical and experimental work on the subject have been recently reviewed by Barbieri *et al.* [10].

The present paper reports the relation between the OGS induced in a uranium-neon hollow-cathode discharge and the laser photon flux, for excitation of ground-state atoms contained in the negative glow of the discharge. It is worth mentioning that the OGS has no regular behaviour in the cathode fall because large variations in the electric-field–to–atomic-density ratio E/N take place there [7, 9]. The relations obtained by solving the corresponding rate equations showed that it is experimentally possible to determine the saturation parameter of the induced transition, that is inversely proportional to the product of the light absorption cross-section and the spontaneous radiative lifetime.

2. – Theory.

It will be assumed that, no matter which process is responsible for the impedance change in the discharge due to light absorption, the magnitude of the optogalvanic signal will be proportional to the upper-level atomic density variation:

$$\Delta V = -A\,\Delta n_{\mathrm{u}}\,. \tag{1}$$

In this equation Δn_{u} is the variation in the upper-level density, A is a positive proportionality factor and ΔV is the magnitude of the optogalvanic signal per unit volume. The minus signal indicates a decrease in the discharge impedance that occurs when atoms in nonmetastable levels are excited by laser light. In order to calculate Δn_{u} it is necessary to solve the rate equations, where the upper level is connected to the ground state (lower level) through radiative and collisional decay in a time T, where $1/T$ is equal to the sum of $1/\tau$ plus $1/\gamma$, τ being the spontaneous radiative lifetime and γ the relaxation time due to collisions between the excited atoms and other species.

Let us consider first that, if there is no laser, the upper level is populated in a characteristic time R by the discharge, as shown in fig. 1*a*).

In a steady state the upper-level population has a constant value, given by

$$n_{\mathrm{u}}(0) = n_{\mathrm{l}}(0)\,\frac{T}{R}\,. \tag{2}$$

If the atoms are illuminated by resonant laser light the population density changes,

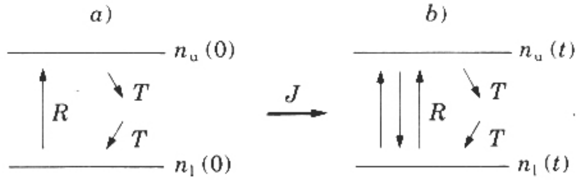

Fig. 1. – Electronic transition processes between two energy levels in an electric discharge. *a*) Without laser; *b*) with resonant laser radiation.

depending on the photon flux J. The rate equations, according to fig. 1b), will be

$$\frac{\mathrm{d}n_\mathrm{u}(t)}{\mathrm{d}t} = n_\mathrm{l}(t)\,\sigma_0\,J - n_\mathrm{u}(t)\,\sigma_0\,J + \frac{n_\mathrm{l}(t)}{R} - \frac{n_\mathrm{u}(t)}{T}\,, \tag{3}$$

$$\frac{\mathrm{d}n_\mathrm{l}(t)}{\mathrm{d}t} = -\frac{\mathrm{d}n_\mathrm{u}(t)}{\mathrm{d}t}\,. \tag{4}$$

This set of differential equations can be solved with the initial conditions that $n_\mathrm{u} = n_\mathrm{u}(0)$ and $n_\mathrm{l} = n_\mathrm{l}(0)$ for $t = 0$. The upper-level population density is then calculated:

$$n_\mathrm{u}(t) = n_\mathrm{u}(0)\exp[-\alpha t] + n\frac{1}{\alpha}\left(\sigma_0\,J + \frac{1}{R}\right)[1 - \exp[-\alpha t]]\,, \tag{5}$$

where $\alpha = 2\sigma_0\,J + 1/R + 1/T$, σ_0 is the absorption cross-section at the centre of the line and $n = n_\mathrm{u}(0) + n_\mathrm{l}(0)$.

For a modulated c.w. laser beam the time of illumination is large compared to the temporal processes involved in the transitions. The exponential factors in eq. (5) go to zero and the upper-level density approaches a constant value:

$$n_\mathrm{u}(\infty) = \frac{1}{\alpha}[n_\mathrm{u}(0) + n_\mathrm{l}(0)]\left(\sigma_0\,J + \frac{1}{R}\right). \tag{6}$$

The density variation $n_\mathrm{u}(\infty) - n_\mathrm{u}(0)$ can be calculated using eqs. (6) and (2):

$$\Delta n_\mathrm{u} = \frac{1}{\alpha}\sigma_0\,J[n_\mathrm{l}(0) - n_\mathrm{u}(0)]\,, \tag{7}$$

so that the OGS can be written as

$$\Delta V = -A\sigma_0\,J\,\Delta n_0\left(2\sigma_0\,J + \frac{1}{R} + \frac{1}{\tau} + \frac{1}{\gamma}\right)^{-1}, \tag{8}$$

where Δn_0 is the difference in the atomic density with the laser turned off. In most atomic transitions the spontaneous radiative lifetime is small compared to the pumping time R and to the relaxation time γ. Neglecting these terms in eq. (8) the OGS can be expressed as

$$\Delta V = -B\frac{1}{1 + \dfrac{S}{J}}\,. \tag{9}$$

In this equation S is defined as the optical saturation parameter [11], that is equal to $1/2\sigma_0\tau$. The proportionality factor B and the parameter S change their values with the discharge current. They can be calculated by taking two points, *e.g.*, the lowest and the highest values of the $\Delta V(J)$ experimental points, and solving a simple system of two algebraic equations.

It can be seen from eq. (9) that for $J \ll S$ the OGS is proportional to $\sigma_0\,J\,\Delta n_0$, that is essentially the same result obtained by Erez *et al.* [5] for the c.w. laser case, where weak absorption was assumed. For strong absorption, $J \gg S$ and the signal approaches asymptotically its maximum value, given by the factor B. At the point where the OGS assumes its half-maximum the photon flux is equal to the saturation parameter S. In this case $\Delta n_u = (1/4)\Delta n_0$. The shape of the curve, showing saturation effects, was also reported in previous papers for c.w. modulated laser beam [9, 12, 13]. It should be noted that the saturation parameter, as defined, can only be determined for lasers whose linewidth $\delta\nu$ is much smaller than the atomic line broadening $\Delta\nu$, otherwise the measured laser power would include considerable amount of photons that do no interact with the absorbing centres.

3. – Experimental.

A diagram of the experimental set-up is shown in fig. 2. The OGS was induced in a home-made water-cooled uranium hollow-cathode lamp. The details of the lamp construction can be seen in the schematic of fig. 3. Two borosilicate glass tubes (Corning 7740) with optical windows of the same material were glued to a water-cooled copper piece with epoxy resin (Henkel Poliamide Resin, Shell Epikote 828). A uranium plug of 5/8 inches in length with an inner bore of 3/16 inches in diameter was placed inside the copper piece, centred on the tube axis. Mica foils were used to prevent discharge on the copper surfaces. A diffusion pump station with a liquid air trap was used in the treatment process of the lamp, before sealing it off. This process of electrode surfaces cleaning is necessary to get a noiseless discharge and it is described elsewhere [14, 15]. The lamp was sealed with 8 Torr neon gas and could be operated with a discharge current up to 200 mA.

A single-mode c.w. dye laser, actively frequency stabilized, with a linewidth of about 1 MHz (Coherent mod. 699/21), tuned to the $0 \rightarrow 16\,900\ \text{cm}^{-1}$ uranium transition was used for the OGS measurements. A single-line Ar^+ laser (Coherent INNOVA 200-15) at the 514.5 nm was used to pump the ring dye laser. The dye used was the R6G. A SPEX 1402 double monochromator was used to position the laser wavelength in the region of interest. A iodine cell was then used as a reference for fine-tuning the laser frequency. The dye-laser mode structure was monitored with a 2 GHz FSR Fabry-Perot interferometer. In the case of mode jump, detected in an oscilloscope, the laser is repositioned in the correct mode of operation and a new record is performed.

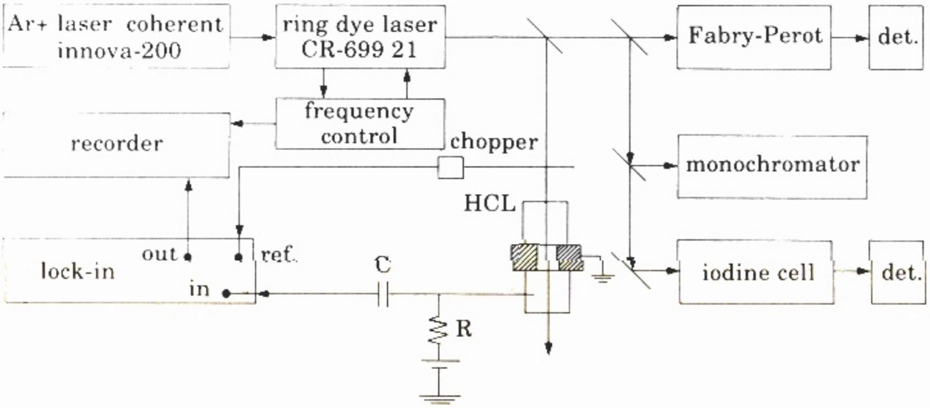

Fig. 2. – Experimental set-up used for the OGS measurements.

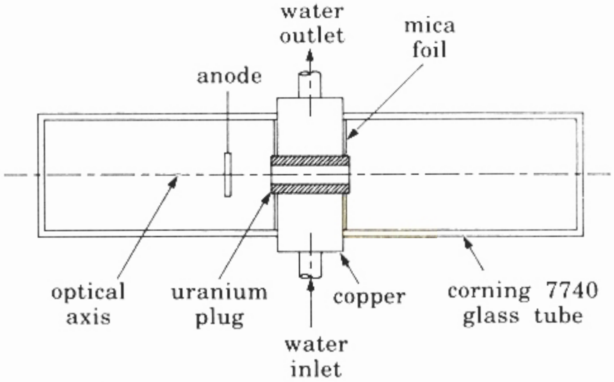

Fig. 3. – Uranium hollow-cathode lamp design.

The laser beam was modulated by a mechanical chopper at 150 Hz and unfocused into the plasma, in order to match the negative glow region, with a diameter of approximately 3 mm. The voltage changes induced by the laser in the lamp were measured through a 1 kΩ ballast resistor and a 0.001 μF capacitor by a lock-in amplifier (Stanford mod. SR530) phase referenced to a mechanical chopper (Stanford mod. SR540). The absorption line profile was obtained by frequency scanning the laser light in a 10 GHz range around the line absorption peak.

4. – Results and comments.

Figure 4 shows the OGS laser power dependence, in arbitrary units, for discharge currents of 60, 100 and 200 mA. The dots are experimental data and the lines represent the calculated shape of the function $\Delta V(P)$. Equation (9) is valid for the laser photon flux J and for the laser power P as well. Therefore, by taking two experimental points on the $\Delta V(P)$ curve and solving the two corresponding algebraic equations, it is easy to calculate the two constants B and S from eq. (9), for each curve. The values of constant B, the laser power saturation P_s, the photon flux saturation S and the $\sigma_0 \tau$ product are shown in table I for the three values of the discharge current.

These results can be compared to the ones measured by optical methods. The

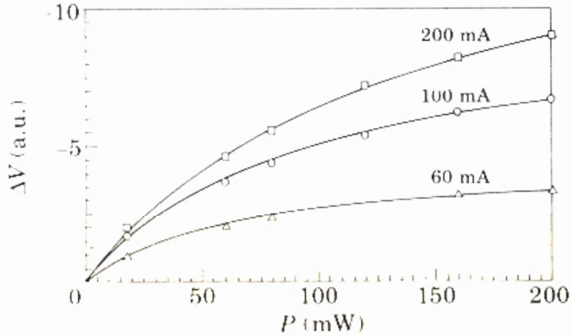

Fig. 4. – OGS as a function of laser power for 60, 100 and 200 mA discharge current. The points are experimental data and the lines are the calculated $\Delta V(P)$ functions.

TABLE I. – *Experimental results.* P_s: *laser power saturation for the* $0 \rightarrow 16\,900\ \text{cm}^{-1}$ *uranium transition;* S: *photon flux saturation (number of photons per unit area per unit time).*

I (mA)	B (a.u.)	P_s (mW)	S ($\text{cm}^{-2}\,\text{s}^{-1}$)	$\sigma_0\,\tau$ ($\text{cm}^2\,\text{s}$)
60	4.4	62	$2.6 \cdot 10^{18}$	$1.9 \cdot 10^{-19}$
100	9.8	92	$3.9 \cdot 10^{18}$	$1.3 \cdot 10^{-19}$
200	14.9	131	$5.6 \cdot 10^{18}$	$0.9 \cdot 10^{-19}$

spontaneous radiative lifetime τ of the upper level is 255 ns, according to Carlson *et al.* [16]. The absorption cross-section σ_0 was measured by Gagné *et al.* [17] in a 2 Torr neon-uranium hollow-cathode tube, with 200 mA discharge current. They have determined σ_0, that is $k(\omega_0)/N$ in table I of their paper, as being equal to $5.8 \cdot 10^{-13}\ \text{cm}^2$. Therefore $\sigma_0\,\tau$ is approximately $1.5 \cdot 10^{-19}\ \text{cm}^2\,\text{s}$, in close agreement with the results presented in this work. The dispersion observed in the $\sigma_0\,\tau$ values for different discharge current can be explained reminding that the peak absorption cross-section is inversely proportional to the Doppler linewidth and so it should decrease with discharge current increase, as can be seen in the last column of table I.

The presented theoretical model does not account for induced transitions between excited levels of species in a hollow-cathode discharge. This condition restricts the practical validity of the derived equations to the OGS measurements for electronic transitions of metal atoms sputtered from the cathode surface. Generally the hollow-cathode tubes are filled with rare gases for sustaining the discharge. The energy gap between the ground state and the first excited state of these atoms is so high that no transition can be stimulated by visible laser radiation. Experimental investigations of the OGS have been carried out in argon [5] and neon discharges [6, 9, 18, 19], where appropriate theoretical models were used to describe the effect. In all these cases, the two levels involved in the induced transitions are connected to many other states through radiative and collisional processes.

* * *

We would like to thank N. Beverini of Università di Pisa for reviewing the manuscript. This work has been partially suported by FAPESP under process no. 90/4128-8.

REFERENCES

- [1] R. B. GREEN, R. A. KELLER, G. G. LUTHER, P. K. SCHENCK and J. C. TRAVIS: *Appl. Phys. Lett.*, **29**, 727 (1976).
- [2] R. A. KELLER and E. F. ZALEWSKI: *Appl. Opt.*, **19**, 3301 (1980).
- [3] C. DRÈZE, Y. DEMERS and J. M. GAGNÉ: *J. Opt. Soc. Am.*, **72**, 912 (1982).
- [4] N. S. KOPEIKA: *Appl. Opt.*, **21**, 3989 (1982).
- [5] G. EREZ, S. LAVI and E. MIRON: *IEEE J. Quantum. Electron*, **QE-15**, 1328 (1979).
- [6] A. BEN-AMAR, G. EREZ and R. SHUKER: *J. Appl. Phys.*, **54**, 3688 (1983).
- [7] E. M. VAN VELDHUIZEN, F. J. DE HOOG and D. C. SCHRAM: *J. Appl. Phys.*, **56**, 2047 (1984).

[8] R. A. KELLER, B. E. WARNER, E. F. ZALEWSKI, P. DYER, R. ENGLEMAN jr. and B. A. PALMER: *J. Phys. (Paris) C*, **7-44**, 23 (1983).
[9] E. F. ZALEWSKI, R. A. KELLER and R. ENGLEMAN jr.: *J. Chem. Phys.*, **70**, 1015 (1979).
[10] B. BARBIERI, N. BEVERINI and A. SASSO: *Rev. Mod. Phys.*, **62**, 603 (1990).
[11] W. DEMTRÖDER: *Laser Spectroscopy* (Springer-Verlag, New York, N.Y., 1981), Chapt. 2.
[12] W. B. BRIDGES: *J. Opt. Soc. Am.*, **68**, 352 (1978).
[13] R. A. KELLER, R. ENGLEMAN jr. and E. F. ZALEWSKI: *J. Opt. Soc. Am.*, **69**, 738 (1979).
[14] B. BARBIERI, N. BEVERINI, M. GALLI, M. INGUSCIO and F. STRUMIA: *Nuovo Cimento D*, **4**, 172 (1984).
[15] W. G. JONES and A. WALSH: *Spectrochim. Acta*, **16**, 249 (1960).
[16] L. R. CARLSON, J. A. PAISNER, E. F. WORDEN, S. A. JOHNSON, C. A. MAY and R. W. SOLARZ: *J. Opt. Soc. Am.*, **66**, 846 (1976).
[17] J. M. GAGNÉ, B. LEBLANC, B. MONGEAU, M. CARLEER and L. BERTRAND: *Appl. Opt.*, **18**, 1084 (1979).
[18] R. SHUKER, A. BEN-AMAR and G. EREZ: *J. Phys. (Paris) C*, **7-44**, 35 (1983).
[19] K. C. SMITH, R. A. KELLER and F. F. CRIM: *Chem. Phys. Lett.*, **55**, 473 (1978).