Oscillator Strength Measurement of Transitions Excited from a Metastable State Using Laser Optogalvanic Spectroscopy - Part 1: Theory

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A theoretical model to determine the oscillator strength of electronic transitions of atoms in a hollow-cathode discharge, excited from a metastable level, was developed. Optogalvanic spectroscopy technique is considered, where the light from a c.w. tunable dye laser stimulates an electronic transition of atoms contained in a hollow-cathode discharge. The rate equations of a three level system are solved analytically and the metastable level density variation is obtained as a function of the laser power. As the optogalvanic signal amplitude (OGS) is proportional to this level density variation, an expression was obtained that relates the OGS to the laser intensity and the oscillator strength of the stimulated transition.

Introduction

It was shown in a previous work [1] that the saturation parameter of an electronic transition of atoms contained in a hollow-cathode discharge could be measured, for transitions excited from the ground state, by using the optogalvanic spectroscopy technique. The principles and applications of this technique are described in a review published by Barbieri et al [2]. The OGS arises whenever the discharge is illuminated by laser light whose frequency is resonant with some electronic transition of atoms present in the plasma. This signal is detected as an impedance change in the electric discharge. The increase in the electron temperature and the direct ionization of atoms excited by the laser light are the main mechanisms that produce the impedance change [3,4,5]. In the first process, part of the laser energy absorbed by the atoms is transferred to the electrons through collisions [3,4]. In the second process, direct ionization of the excited atoms by electron collisions may occur, that produces the impedance change in the discharge [5]. It is assumed that the magnitude of the signal is proportional to the variation of the atomic level densities due to the laser light absorption, no matter which mechanism may induce the impedance change. However, signals of opposite polarities are recorded in an usual optogalvanic spectrocopy experiment. While excitation of resonant levels produces a decrease in the plasma resistivity, excitation of metastable ones has an inverse effect, increasing the plasma resistivity. This can be explained by considering the fact that metastable atoms play a fundamental rule in sustaining the electrical discharge. The long lifetime and the high energy levels of these atoms are a favorable source for atomic ionization through the various collisional processes that may occur in the plasma, so that depopulation of these levels by laser light will increase the plasma resistivity.

Bearing in mind that it is not possible to establish a complete relationship between the OGS magnitude and the several parameters involved, such as electron density and temperature, current density, light absorption cross section and others, a simplified theoretical model was developed, where the only variable parameter is the laser light intensity. A further simplification to get an analytical solution for the rate equations was to stimulate electronic transition where the lower level is a metastable one. In this case, depopulation of these levels by spontaneous decay is neglected, compared to the density time variation of the other levels involved.

Theory

In the case where the laser light stimulates an electronic transition from a metastable state in a hollow-cathode discharge, the optogalvanic signal magnitude will be assumed to be proportional to the density variation of this level, that is:

(1)
$$\Delta V = A \cdot \Delta n_2$$

In this equation Δn_2 is the metastable level variation produced by the laser-atom interaction, A is a proportionality factor and ΔV is the optogalvanic signal amplitude per unit volume. A three level system is used to calculate Δn_2. The rate equations describe the density variations of the levels from $t = 0$, when the laser is just turned on, to a time long enough $(t = \infty)$, when the system photons-atoms attains the equilibrium and $\frac{dn_i}{dt} = 0$. In this work a c.w. laser modulated at 37 Hz by a mechanical chopper was used for the electronic excitation. As the "laser on" time is much longer than the temporal processes involved in the electronic transitions, the condition of steady state will be assumed for the final density values of the atomic levels. The rate equations, according to figure 1, are:

(2)
$$\frac{dn_1}{dt} = -\frac{n_1}{R_{13}} - \frac{n_1}{R_{12}} + \frac{n_3}{\tau_{31}} + \frac{n_2}{R_{21}}$$

(3)
$$\frac{dn_2}{dt} = \frac{n_1}{R_{12}} - \frac{n_2}{R_{21}} - \frac{n_2}{R_{23}} + \frac{n_3}{\tau_{32}} - n_2\sigma_0 J + n_3\sigma_0 J$$

(4)
$$\frac{dn_3}{dt} = \frac{n_1}{R_{13}} + \frac{n_2}{R_{23}} - \frac{n_3}{\tau} + n_2\sigma_0 J - n_3\sigma_0 J$$

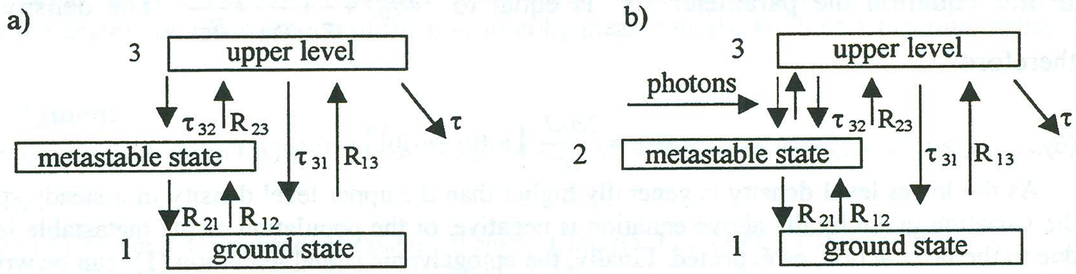

Fig.1: Schematic diagram of the main electronic transition processes for population changes of a three level system in a hollow-cathode discharge. **R** are the relaxation times of density changes due to collisions and τ are the spontaneous radiative lifetimes. **a)** without laser; **b)** with resonant laser radiation tuned to the $2 \rightarrow 3$ transition.

In these equations σ_0 is the light absorption cross section at the line center, J is the photon flux, R are the relaxation times due to collisions and τ are the spontaneous radiative lifetimes of the levels. The units used for the two first parameters in this work are cm^2 and $cm^{-2} \cdot s^{-1}$, respectively. The density variation of the metastable level due to radiative decay was neglected. The absolute values of the n_i are irrelevant and, in fact, they must be normalized to their respective degeneracy. The aim of solving these equations is to find out the value of n_2 at time long enough for the equilibrium conditions to be attained. This is a fundamental condition to calculate the variation Δn_2, that is equal to $n_2(\infty) - n_2(0)$. If the "laser on" time is too short, as in the case of pulsed lasers, the solution for $n_i(t)$ contains the factor $\left(1 - e^{-\beta t}\right)$, where β is a parameter that depends on the product $\sigma_0 J$ and the inverse of the relaxation times. Such solution can be found in ref. 1 and was obtained by solving the rate equations of a more simple two level system.

The initial conditions for $t = 0$ ($J = 0$ and $\frac{dn_i}{dt} = 0$) give some relations between the density of the levels considered in the rate equations. The value of $n_2(\infty)$ can be determined from the equations 3 and 4 taking into account the relations of the initial conditions and the following assumptions and approximations:

a) $\frac{n_2(0)}{R_{23}} << \frac{n_1(0)}{R_{13}}$ and $\frac{n_2(0)}{R_{23}} << \frac{n_2(0)}{R_{21}}$

b) $\sum_{i=1}^{3} n_i(t) = n$ at any time.

c) $n_1(0) = n_1(\infty)$ - (The laser interaction with the metastable level does not introduce appreciable changes in the ground state density)

d) $n_3(\infty) = \sum_{i=1}^{3} n_i(0) - \sum_{i=1}^{2} n_i(\infty)$

Considering such approximations by means of physical arguments is an usual procedure to solve analytically multilevel rate equations [6]. The result for the metastable level density is:

$$n_2(\infty) = \frac{1}{\alpha} \cdot \left[2\sigma_0 J n_2(0) + 2\sigma_0 J n_3(0) + n_2(0) \cdot \left(\frac{1}{\tau} + \frac{1}{\tau_{32}} + \frac{1}{R_{21}} \right) \right] \quad (5)$$

In this equation the parameter α is equal to $4\sigma_0 J + \frac{1}{\tau} + \frac{1}{\tau_{32}} + \frac{1}{R_{21}}$. The density variation is therefore:

$$\Delta n_2 = \frac{2\sigma_0 J}{\alpha} \cdot [n_3(0) - n_2(0)] \quad (6)$$

As the lower level density is generally higher than the upper level density in a steady state discharge, the variation given by the above equation is negative, or the population of the metastable level decreases due to the laser action, as expected. Finally, the optogalvanic signal -equation (1)- can be written as

$$\Delta V = B \cdot \left(1 + {J_s}/{J} \right)^{-1} \quad (7)$$

The parameter J_s is defined as the saturation photon flux and is given by:

$$J_s = \frac{1}{4\sigma_0} \cdot \left(\frac{1}{R_{21}} + \frac{1}{\tau} + \frac{1}{\tau_{32}} \right) \quad (8)$$

The equation (7) shows that for low laser light intensity, i.e. $J << J_s$, the signal amplitude is proportional to $\sigma_0 J[n_2(0) - n_3(0)]$, that agrees with the result obtained by Erez *et al.*[7]. If $J >> J_s$, that is the case of strong absorption, the signal amplitude approaches asymptotically to its maximum value, given by the factor B. The values of this factor and of the saturation photon flux will depend on the hollow cathode discharge conditions. They were found by fitting the equation (7) to the OGS amplitude, measured at the line center, as a function of the laser power. To determine the saturation parameter using this technique a condition that the laser linewidth be much smaller than the Doppler broadening must be fulfilled, otherwise part of the measured laser power should correspond to photons that do not interact with the group of atoms whose velocity are perpendicular to the laser beam direction. For low frequency laser modulation $\tau \ll \tau_{32} \ll R_{21}$ and the equation (8) can be written as $J_s = (4\sigma_0\tau)^{-1}$. The absorption cross section is proportional to the oscillator strength through the relation:

(9) $$f_{23} = \frac{mc \cdot \Delta\nu_D \cdot \sigma_0}{e^2 \cdot 2\sqrt{\pi \ln 2}}$$

In this equation m and e are the electron mass and electron charge, c is the light velocity in the medium and $\Delta\nu_D$ is the Doppler broadening of the absorption line. Finally, the oscillator strength can be written in terms of $\Delta\nu_D$, J_s and τ, using the equation (9) and the relation between σ_0, J_s and τ.

(10) $$f_{23} = 10 \cdot \frac{\Delta\nu_D}{J_s \tau}$$

By scanning the laser frequency around the line absorption center, the Doppler broadening can be measured directly from the line shape of the optogalvanic signal. In this case, low laser power must be used in order to avoid saturation effects. From the experimental data of the OGS as a function of the laser power it is possible to determine J_s by fitting the equation (7) to these experimental measurements.

Conclusion

A theoretical model was developed that relates the optogalvanic signal amplitude to the saturation parameter of an electronic transition excited from a metastable state. It was shown that, once the total radiative lifetime of the upper level is known, it is possible to determine experimentally the oscillator strength or the absorption cross section of the transition by measuring the OGS as a function of the laser power.

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References

[1] - A.Mirage, D.Pereira, F.C.Cruz and A.Scalabrin, Il Nuovo Cimento, 14 D (1992) 605.
[2] - B.Barbieri, N.Beverini and A.Sasso, Rev. Mod. Phys. 70 (1990) 603.
[3] - R.A. Keller and E.F. Zalewski, Appl. Opt. 19 (1980) 3301.
[4] - C. Drèze, Y. Demers and J.M. Gagné, J. Opt. Soc. Am. 72 (1982) 912.
[5] - N.S. Kopeika, Appl. Opt. 21 (1982) 3989.
[6] - A.E. Siegman, Lasers (University Science Books, Mill Valley, 1986) ch.4.
[7] - G.Erez, S.Lavi and E.Miron, IEEE J.Quantum Electron. QE-15 (1979) 1328