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Saturation measurements of the a $^3P_0$–y $^3D_1^o$ Ti I transition by optogalvanic spectroscopy

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Abstract

The optogalvanic signal (OGS) induced in a Ti–Ar hollow-cathode discharge, stimulated by modulated cw laser light tuned to the a $^3P_0$–y $^3D_1^o$ Ti I transition, was measured as a function of the light intensity. By solving analytically the rate equations of a three-level system, which include the Ti I ground state, the lower metastable a $^3P_0$ and the upper y $^3D_1^o$ levels, an expression was obtained that relates the magnitude of the OGS to the laser intensity and the saturation parameter $(1/\sigma_0\tau)$. The $\sigma_0\tau$ product was determined by fitting the theoretical curve to the experimental data. © 1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

In a previous work [1] it was shown that the optical saturation parameter of an electronic transition of species contained in a hollow-cathode discharge could be measured, for some particular experimental conditions, by using the well-known optogalvanic spectroscopy technique. The principles and applications of this technique are described in a review published by Barbieri et al. [2]. The optogalvanic signal (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 of electron temperature and the direct ionization of atoms excited by the laser light are the main mechanisms that produce the impedance change [3–5]. 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, which 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 laser light absorption, no matter which mechanism may induce the impedance change. However, signals of opposite polarities are recorded in a usual optogalvanic spectroscopy experiment. While excitation of resonant levels produces a decrease of the plasma resistivity, excitation of metastable ones has an inverse effect, increasing the plasma resistivity. This can be explained by the fact that the metastable atoms play a fundamental role 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.

As 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 obtain an analytical solution of the rate

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equations was to consider a stimulated 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.

2. Theory

When 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:

$$\Delta V = A \Delta n_2 . \tag{1}$$

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 equilibrium and $\mathrm{d}n_i/\mathrm{d}t = 0$. In this work a cw 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 Fig. 1, are:

$$\frac{\mathrm{d}n_1}{\mathrm{d}t} = -\frac{n_1}{R_{13}} - \frac{n_1}{R_{12}} + \frac{n_3}{\tau_{31}} + \frac{n_2}{R_{21}}, \tag{2}$$

$$\frac{\mathrm{d}n_2}{\mathrm{d}t} = \frac{n_1}{R_{12}} - \frac{n_2}{R_{21}} - \frac{n_2}{R_{23}} + \frac{n_3}{\tau_{32}} - n_2\sigma_0 J + n_3\sigma_0 J, \tag{3}$$

$$\frac{\mathrm{d}n_3}{\mathrm{d}t} = \frac{n_1}{R_{13}} + \frac{n_2}{R_{23}} - \frac{n_3}{\tau} + n_2\sigma_0 J - n_3\sigma_0 J. \tag{4}$$

In these equations σ_0 is the light absorption cross section at the line centre, 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 $\mathrm{cm}^{-2}\ \mathrm{s}^{-1}$, respectively. The density variation of the metastable level due to radiative decay was neglected. The absolute values of n_i are irrelevant and, in fact they must be normalized by their respective degeneracy. The aim of solving these equations is to find the value of n_2 at time long enough such that the equilibrium conditions had already been attained. This is a fundamental condition to calculate the variation Δn_2 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 $1 - \exp(-\beta t)$, where β is a parameter that depends on the product $\sigma_0 J$ and the inverse of the relaxation times. Such a solution can be found in Ref. [1] and was obtained by solving the rate equations of a simpler two-level system.

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

1. $n_2(0)/R_{23} \ll n_1(0)/R_{13}$ and $n_2(0)/R_{23} \ll n_2(0)/R_{21}$.
2. $\Sigma_{i=1}^{3} n_i(t) = n$ at any time.
3. $n_1(0) = n_1(\infty)$. (The laser interaction with the metastable level does not introduce appreciable changes in the ground state density.)
4. $n_3(\infty) = \Sigma_{i=1}^{3} n_i(0) - \Sigma_{i=1}^{2} n_i(\infty)$.

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

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

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

$$\Delta n_2 = \frac{2\sigma_0 J}{\alpha}\left[n_3(0) - n_2(0)\right]. \tag{6}$$

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, that is, the population of the metastable level decreases due to the laser action, as expected.

Finally, the optogalvanic signal – Eq. (1) – can be written as:

$$\Delta V = B\left(1 + \frac{J_S}{J}\right)^{-1}. \tag{7}$$

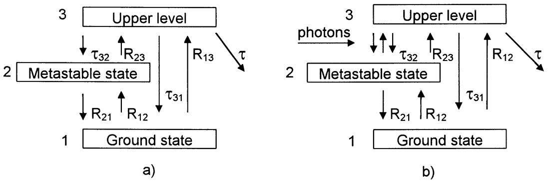

Fig. 1. Schematic diagram of the main electronic transition processes for population changes of a three level system in a hollow-cathode discharge. R_{ij} are the relaxation times of density changes due to collisions and τ_{ij} are the spontaneous radiative lifetimes. (a) Without laser; (b) with resonant laser radiation tuned to the 2 → 3 transition.

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

$$J_S = \frac{1}{4\sigma_0}\left(\frac{1}{\tau} + \frac{1}{R_{21}} + \frac{1}{\tau_{32}}\right). \tag{8}$$

Eq. (7) shows that for low laser light intensity, i.e. $J \ll J_S$, the signal amplitude is proportional to $\sigma_0 J[n_2(0) - n_3(0)]$, which agrees with the result obtained by Erez et al. [7]. If $J \gg J_S$, i.e. for strong absorption, the signal amplitude approaches asymptotically 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 Eq. (7) to the OGS amplitude, measured at the line centre, as a function of the laser power (Section 4). To determine the saturation parameter using this technique, the condition of laser linewidth much smaller than de 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 is perpendicular to the laser beam direction.

3. Experimental

A schematic diagram of the experimental set-up for the OGS measurements is shown in Fig. 2. The signal was induced in a titanium–argon home-made, water-cooled, hollow cathode tube. The geometry and the process of tube construction are similar to that described in Ref. [1], except for the cathode element, that was replaced by a titanium plug. This plug was a cylindrical piece 23 mm long with a see-through hole of 3.2 mm in diameter, drilled on its axis. The tube was sealed with 2.5 Torr argon gas and was operated with a 150 mA stabilized dc discharge.

A ring dye laser (Spectra-Physics model 380A), with about 20 MHz linewidth, was tuned to the a $^3P_0$–y $^3D_1^o$ Ti I transition in the 592.2 nm wavelength region. The lower level transition is a metastable one with energy of 8436.63 cm^{-1}. The upper level (25317.842 cm^{-1}) is strongly coupled by allowed dipole transitions to the ground state (a$^3F_2$), as well as to many other intermediate levels. A multi-line Ar$^+$ laser (Spectra Physics model 171) was used to pump the dye laser that operated with a Rhodamine 6G solution. The tuning and scanning of the laser wavelength was monitored by a wavemeter (Burleigh model WA-1000) with a precision of 0.001 nm. The entire scanning in the selected frequency range and some eventual mode jumps could be observed on the wavemeter display.

The laser beam, that was modulated by a mechanical chopper (Stanford model SR540) at 37 Hz, matched the negative glow inside the cathode hole, with a diameter of about 3 mm. The optogalvanic signal induced by the laser was measured through a ballast resistor (500 Ω) and a dc decoupling capacitor (1 pF) by a lock-in amplifier (Stanford model SR510), phase referenced to the chopper. The lock-in output signal was digitized and connected to a PC through a GPIB interface for storage and analysis.

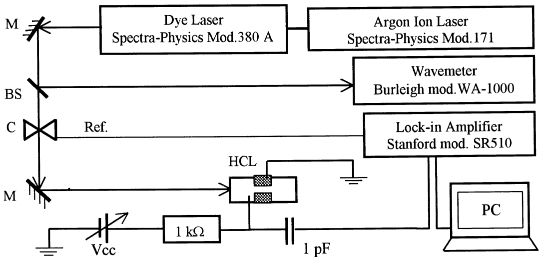

Fig. 2. Experimental set-up used for optogalvanic signal detection. M – mirrors, BS – beam-splitter, C – mechanical chopper, HCL – hollow-cathode lamp.

4. Results and comments

The results of the OGS measurements are shown in Fig. 3, where the dots correspond to the experimental data and the solid line are the fitting of ΔV as a function of the laser power. The OGS amplitude, in arbitrary units scale, was measured at the absorption line peak by scanning the laser frequency in a 10 GHz range around this peak. By fitting Eq. (7) to the experimental data the values of B and P_S where obtained, $B = 35$ and $P_S = 700$ mW, where P_S is the laser power saturation, for a 150 mA discharge current. As the area of the laser beam cross section is approximately 0.07 cm^2 and the photon energy is 2.09 eV, the saturation photon flux was estimated to be 3.0×10^{19} photons cm^{-2} s^{-1}. This value can be compared with that obtained from Eq. (8), using the numerical values of τ and τ_{32} from Refs. [8,9]. The total lifetime τ was measured by Salih and Lawler [8] and is equal to 14.5 ns. The τ_{32} lifetime can be calculated from the f_{23} value found in Ref.

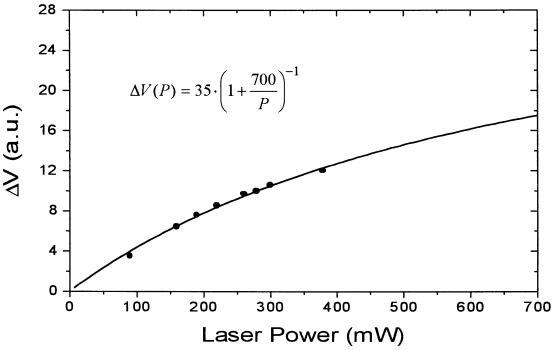

Fig. 3. OGS as a function of laser power measured at the absorption line centre, for 150 mA HCD current operation. The points are experimental data and the solid line is the fitting of the function $\Delta V(P)$.

[9], where f_{23} is the oscillator strength of the transition, equal to 0.0342. Therefore τ_{32} is equal to 460 ns, so that the terms $1/\tau_{32}$ and $1/R_{21}$ can be neglected in Eq. (8), compared to the term $1/\tau$. Eq. (8) is reduced to $J_S = 1/4\sigma_0\tau$. The absorption cross section σ_0 is calculated from Eq. (9), below, where $\Delta\nu_D$ is the Doppler broadening of the absorption line [10], e and m are the electron charge and electron mass and c is the velocity of light in the medium,

$$\sigma_0 = \frac{2}{\Delta\nu_\mathrm{D}}\sqrt{\pi\ln 2}\,\frac{e^2 f_{23}}{mc}. \tag{9}$$

The Doppler broadening measured from the absorption line profile was 1.4 GHz (see also Ref. [11]). Using these microscopic parameters the absorption cross section at the line peak is $\sigma_0 = 6.1\times10^{-13}$ cm^2 and the saturation photon flux is therefore $J_\mathrm{S} = 2.8\times10^{19}$ cm^{-2} s^{-1}, which is in good agreement with the value obtained experimentally in this work.

It was shown that, under particular experimental conditions and in the case where a metastable level is excited by laser light, it is possible to measure the saturation parameter of the transition using optogalvanic spectroscopy techniques. No attempt has been made to determine the accuracy of such measurements, but the deviations that might occur on these experimental observations are mainly due to errors introduced by the measurements of the laser power and the laser beam diameter.

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