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Abrupt changes in neon discharge plasma detected via the optogalvanic effect

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ABSTRACT

When a laser is tuned between two excited energy levels of a gas in a Direct Current discharge lamp, the discharge current will experience a temporary disturbance lasting tens or hundreds of microseconds known as the optogalvanic effect. We have carried out extensive studies of optogalvanic effects in neon discharge plasmas for transitions at 621.7 nm, 630.5 nm, 638.3 nm, 650.7 nm and 659.9 nm. A nonlinear least-squares Monte Carlo technique has been used to determine the relevant amplitude coefficients, decay rates and the instrumental time constant. We discovered an abrupt change in the neon discharge plasma at a discharge current of about 6 mA.

1. Introduction

Laser optogalvanic spectroscopy is a sensitive technique that can be used for a variety of spectroscopic studies, including plasma diagnostics and analytical flame spectroscopy [1]. The optogalvanic effect takes place because of the impedance change in a discharged plasma tube when it is irradiated with laser radiation of a specific wavelength [2]. We have developed a mathematical model and a nonlinear least-squares Monte Carlo algorithm for fitting the optogalvanic signal waveforms associated with the neon transitions for a variety of current values spanning the range 2–19 mA [3–5]. The optogalvanic signal can be described by the sum of a series of exponential functions with the amplitudes and decay rates treated as adjustable parameters when fitted to the experimental signals.

2. Experimental details

A pulsed laser (of typical pulse width 5 ns) was tuned to the appropriate wavelength (621.7 nm, 630.5 nm, 638.3 nm, 650.7 nm, 659.9 nm, corresponding to $1s_5$–$2p_8$, $1s_4$–$2p_6$, $1s_4$–$2p_7$, $1s_4$–$2p_8$, $1s_2$–$2p_2$ transitions, respectively) and directed to enter a hollow cathode discharge lamp (the galvatron) containing neon gas. The galvatron is coupled in series with a Resistor–Capacitor circuit, and the discharge current (in the range 2–19 mA) was controlled by changing the voltage on a power supply (see Fig. 1) [3].

The deviation of the discharge current from its steady state value is the optogalvanic signal, which was displayed on a digital oscilloscope, and averaged over 256 laser pulses. The stored optogalvanic data from the oscilloscope was converted to ASCII format and analyzed using a nonlinear least-squares Monte Carlo fitting routine.

3. Analyses, results and discussion

The mechanism of the optogalvanic effect is best described in the energy diagram in Fig. 2. A laser excites molecules from level L_1 to L_2. Frequent collisions transfer molecules from L_2 to L_2'. Subsequently, molecules radiatively decay down to lower levels L_3, L_4 and L_5, achieving a different population distribution. The new population distribution will have a total ionization rate that produces a different

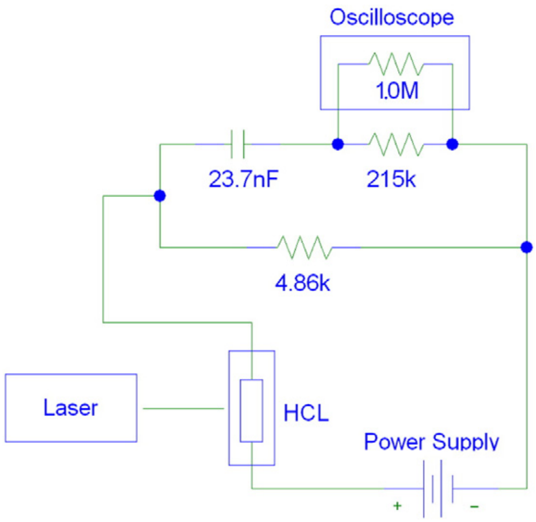

Fig. 1. Experimental arrangement for laser optogalvanic spectroscopy.

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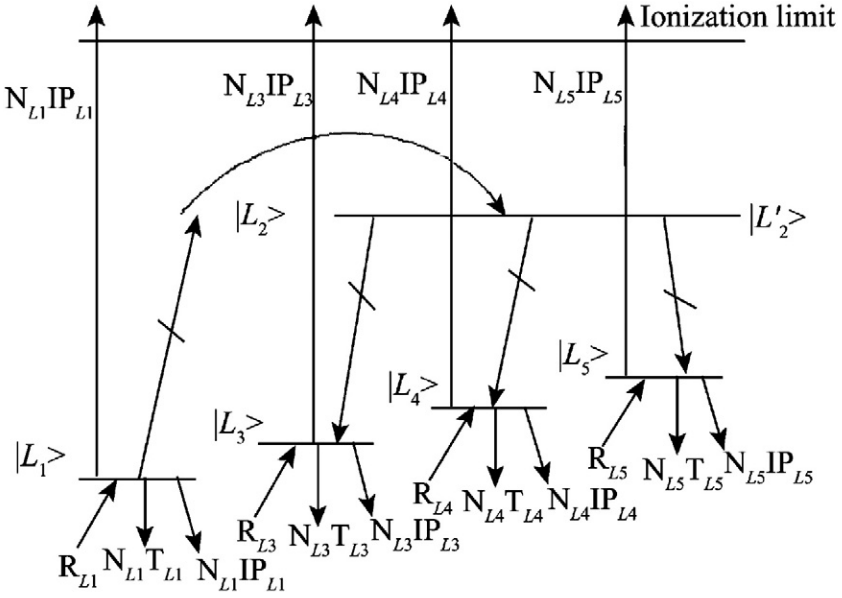

Fig. 2. Energy-level diagram illustrating the optogalvanic effect.

discharge current. The optogalvanic effect signal can be described by the sum of exponential functions given by [1–4]:

$$S(t)=\sum_{j=1}^{j_{max}}\frac{a_j}{1-b_j}\left[\exp\left(-b_j t\right)-\exp\left(-{}^{t}/_{\tau}\right)\right]. \tag{1}$$

The summation is over the lower states relevant to the optogalvanic effect. For this study, they are the four 1s states ($1s_2$, $1s_3$, $1s_4$ and $1s_5$). The exponential rate b_j is given by:

$$b_j=\Gamma_j+I*P_j, \tag{2}$$

where I is the discharge current, Γ_j is related to the spontaneous decay rate for the state j, and P_j is related to the collisional excitation/de-excitation and ionization cross-sections for the state j [1]. In principle, the summation in Eq. (1) should sum over all four 1S states. However, in most cases, not all of the four states are equally important and involved in a particular optogalvanic effect transition. As a result, it is common that we can fit the experimental signal using less than four terms in Eq. (1) for a particular optogalvanic effect signal, and the minimum will involve two states.

The stored optogalvanic data of the 621.7 nm for currents from 2 mA–19 mA was fitted to Eq. (1) using the Monte Carlo fitting routine in order to extract parameters {τ, a_j, b_j} [5] to extract the exponential rates {b_j}. Fig. 3 shows the observed and the fitted curve of the 10 mA optogalvanic signal of 621.7 nm.

Fig. 4 shows the exponential decay rates versus the currents from 2 mA to19 mA for the transition $1s_5$–$2p_8$ transition at 621.7 nm. As seen from Figs. 4–5, the exponential rates, b_j, do not change significantly as the discharge current decreases from 19 to 8 mA. However, for 6 and 7 mA, the exponential rates suddenly become very small.

The values of the decay constants b_2 of the initial states of the five transitions 621.7 nm (s_5), 630.5 nm (s_4), 638.3 nm (s_4), 650.7 nm (s_4) and 659.9 nm (s_2) are shown in Fig. 5. From the figure it is very clear that the decay constant shows abrupt changes for smaller currents and shows a smooth variation from currents 10–19 mA. The same pattern is also observed for the decay constants b_1 and b_3 that are obtained due to excited neon atoms relaxing to optically allowed transitions for all the above mentioned five transitions.

4. Conclusions

In the present work we were able to show that the decay rate constants obtained for the initial states as well as optically allowed states changes abruptly for the smaller values of the current from 2 to 9 mA

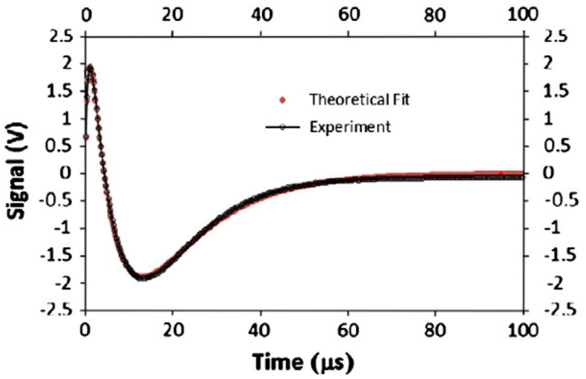

Fig. 3. Observed and fitted optogalvanic waveforms of the 621.7 nm neon transition for 10 mA current.

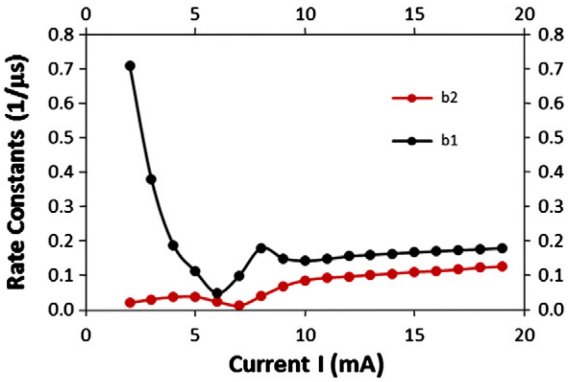

Fig. 4. A plot of the exponential rates versus current of the 621.7 nm waveform.

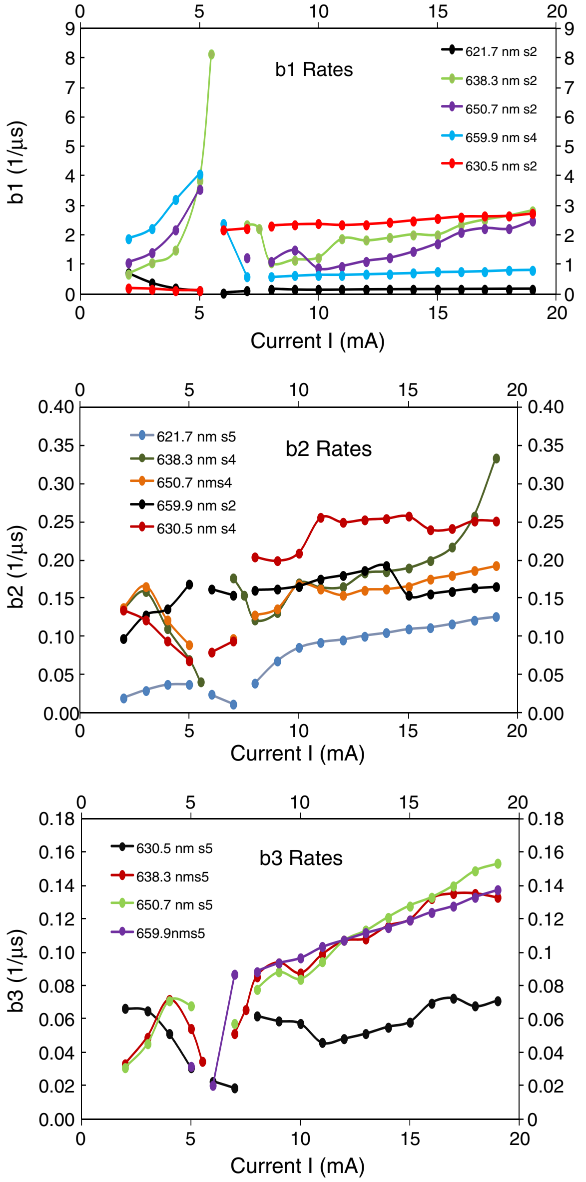

Fig. 5. Variation of the b_1 rate constant as a function of current for the five (5) neon transitions.

and a smooth change for currents from 10 to 19 mA. We also observed a strong indication of state-dependence of the optogalvanic signals. The five optogalvanic transitions of neon investigated in this study can be grouped state-wise as follows: $1s_5$–$2p_8$ (621.7 nm), $1s_4$–$2p_{6,7,8}$ (630.5 nm, 638.3 nm, 650.7 nm) and $1s_2$–$2p_2$ (659.9 nm). The optogalvanic signal amplitudes are a function of the population difference between the states and the ionization rates of the states involved, and the rate constants are governed by the nature of the relaxation of the excited neon atoms to lower energy levels and followed by optically allowed transitions. At higher discharge currents (8–19 mA), one expects enhanced ionization associated with the upper states, and thereby the discharge current variation influences the signal amplitude via the ionization rate rather than the change in population distribution. For lower discharge currents (2–7 mA), the population difference in the energy states is relatively higher, and thereby the laser pulse incident on the neon plasma brings about a larger disturbance in the state-dependent population distribution and induces the abrupt changes noted around 6–7 mA in the rate constant variation associated with the plasma. According to Eq. (2), the abrupt change in optogalvanic signal around $I = 6$ mA is indicative that P_j experiences resonance effects. Since P_j is related to collision cross-sections, it appears that the plasma dynamics in the discharge environment creates an electron energy distribution that induces a collisional resonance, which should be worth further theoretical explorations.

Acknowledgments

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