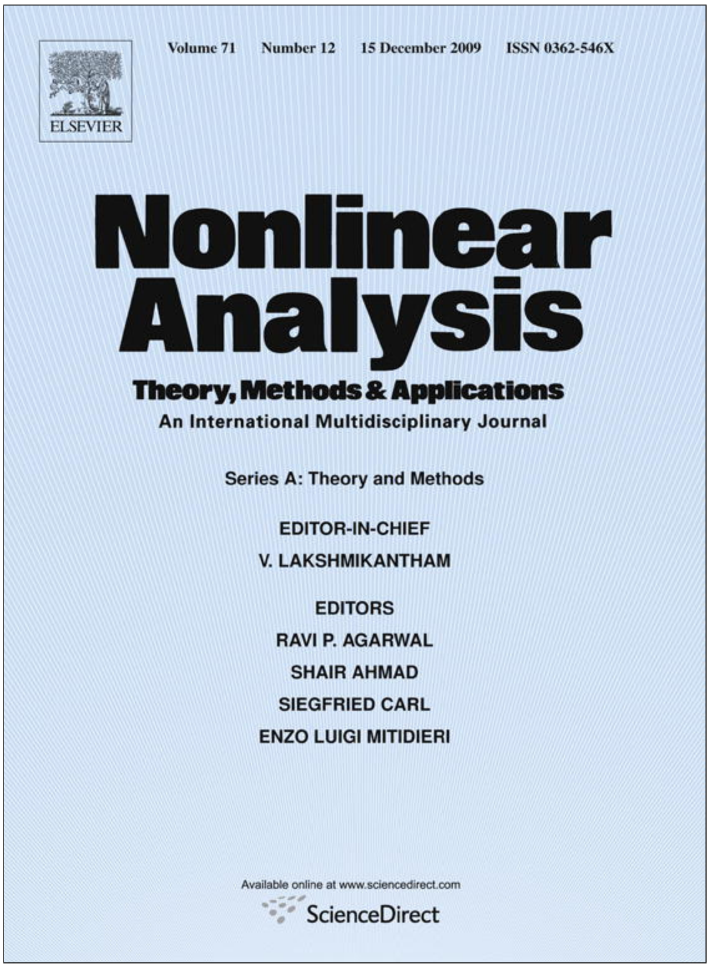

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Laser optogalvanic spectroscopy of neon at 659.9 nm in a discharge plasma and nonlinear least-squares fitting of associated waveforms

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ABSTRACT

Laser optogalvanic spectroscopy of neon has been studied in the discharge environment of a hollow cathode discharge lamp. We illustrate the fitting of the time-resolved optogalvanic waveforms by focusing on the $1s_2$–$2p_2$ transition of neon at 659.9 nm and employ a nonlinear least-squares Monte Carlo approach to determine the pertinent amplitude coefficients, decay rates and the instrumental time constant.

1. Introduction

Laser optogalvanic spectroscopy is a technique for observing the OptoGalvanic (OG) effect associated with an atomic species in a plasma environment utilizing an appropriately tuned laser and pertinent electronics for spectral data acquisition [1,2]. The OG effect occurs due to the impedance change in a discharge tube when it is illuminated with radiation of a specific frequency. Earlier research done by our group has provided libraries of optogalvanic transitions for neon in the visible (590–670 nm) [1] and for argon in the UV (300–325 nm) [3]. We have also utilized the Monte Carlo least-squares fitting method [4,5] to generate an accurate equation for modeling, and have developed a stable fitting algorithm to provide a better understanding of optogalvanic waveforms. In this paper, we present the Monte Carlo fitting of the signal waveform associated with the $1s_2$–$2p_2$ neon transition at 659.9 nm for a variety of current values in the range 2–19 mA.

2. Experimental

A pulsed laser (of typical pulse width 5 ns) is tuned to the appropriate wavelength (659.9 nm) and directed to enter a hollow cathode discharge lamp ("the galvatron" HCL) filled with neon gas. The galvatron is coupled in series with a current-limiting *RC* circuit, and the discharge current (2–19 mA) is controlled by adjusting the voltage on the power supply. The resulting optogalvanic signal (deviation of the discharge current from its steady state value) is displayed on a digital oscilloscope (Tektronix TDS 224; Input Impedance $= 1\ \mathrm{M\Omega}$) and averaged over 256 pulses. The stored data set from the oscilloscope is subsequently converted to DOS text or ASCII format and further analyses performed using a nonlinear least-squares fitting program [4,5].

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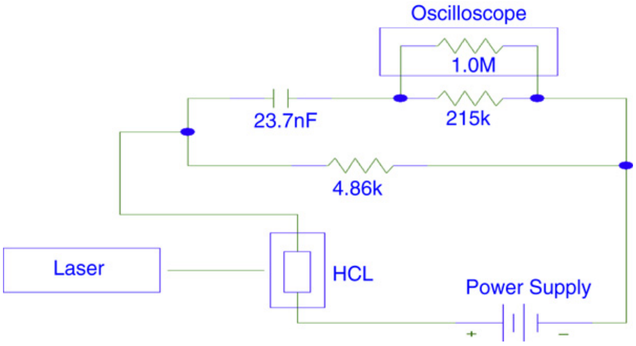

Fig. 1. Experimental arrangement for laser optogalvanic spectroscopy.

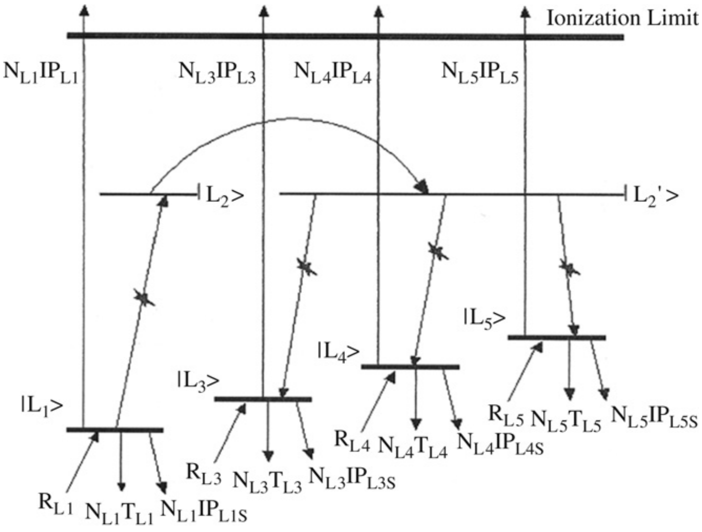

Fig. 2. Simplified energy-level diagram illustrating the optogalvanic transitions in neon.

3. Results and discussion

Our OG spectral data acquisition and least-squares fitting approach has focused on the following: (i) recording and analysis of the laser optogalvanic spectra and waveforms for neon; (ii) refinement of the mathematical rate equation model developed in our laboratory, incorporating the various processes contributing to the generation of an optogalvanic signal in a discharge plasma; (iii) determination of the amplitudes, decay rates and the appropriate instrumental time constant using a nonlinear least-squares fit of the observed time-resolved optogalvanic waveforms; and (iv) development and use of a robust and stable Monte Carlo least-squares fitting technique in conjunction with the mathematical rate equation model to simulate the optogalvanic waveforms optimally.

Fig. 2 given below is a simplified energy-level diagram illustrating the dynamics involved in the generation of the optogalvanic signal. Neon atoms are detected by the OG effect (via enhanced or diminished discharge current in the circuit of Fig. 1) as they undergo ionization. When the laser is in resonance with a specific transition between the energy levels, the signal intensity is primarily governed by the following three processes: (i) electron collisional excitation, (ii) radiative depopulation, and (iii) electron collisional ionization. The theoretical model describing the formation of the time-dependent OG signal is given in detail in a previous publication from our research group [3]. The laser excites the atoms from level L_1 to L_2; following this, frequent collisions transfer the atoms from L_2 to L_2' (as shown in Fig. 2). Subsequently, the neon atoms radiatively decay to lower levels L_3, L_4, and L_5, and acquire a new population distribution, which in turn results in a total ionization rate that generates a different discharge current in the circuit of Fig. 1.

For the neon 1s–2p transition, employing the Paschen notation, there are four 1s states and ten 2p states, which produce a total of 30 allowed radiative transitions. Of the four 1s states, two ($1s_3$ and $1s_5$) are metastable, whereas the other pair ($1s_2$ and $1s_4$) decay to the ground state rapidly within a few ns. In the present paper, we choose to record and analyze the $1s_2$–$2p_2$ transition that occurs at 659.9 nm. The time-resolved experimental OG waveforms recorded were fitted to the following equation:

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

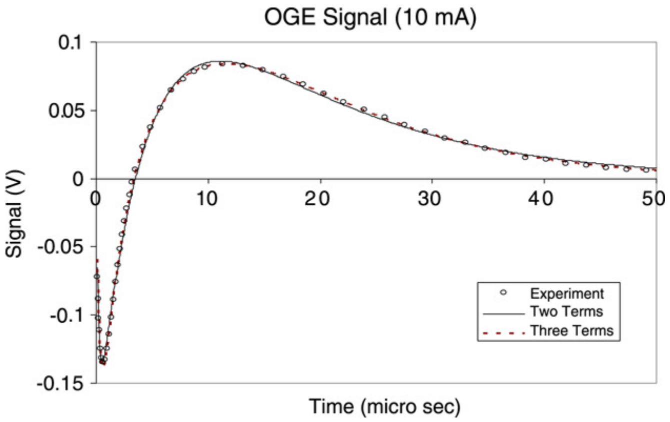

Fig. 3. Comparison of the experimental OG waveform with the theoretically fitted shapes obtained using two and three terms in the rate equation model summarized by Eq. (1).

Table 1
Fitted amplitudes, decay rates and time constant obtained from the nonlinear least-squares fit of the observed OG waveform for neon at 659.9 nm for a current of 10 mA, using two and three terms, respectively, in Eq. (1).

Parameter	Two-term fitted values	Three-term fitted values
τ (µs)	0.206	0.274
a_1 (V)	0.461	−0.113
b_1 (μs^{-1})	0.217	0.647
a_2 (V)	0.288	−0.980
b_2 (μs^{-1})	0.0731	0.134
a_3 (V)		0.894
b_3 (μs^{-1})		0.0966

where the amplitudes a_j and the rates b_j are governed by collisions occurring within the plasma of the hollow cathode discharge lamp, and τ is an instrumental time constant that combines the effects of the plasma relaxation in the discharge and the response of the electrical circuit used to generate the waveforms.

Fig. 3 below shows the quality of the fit obtained between the experimental and theoretically fitted data employing Eq. (1). We have compared the fittings with two ($j = 1$ and 2) and three terms ($j = 1$, 2 and 3) using Eq. (1) and the results are illustrated in Fig. 3. We note that our Monte Carlo approach [4,5] allows us to fit three terms simultaneously without causing any instability, in spite of the fitting parameters being highly correlated, whereas using our earlier traditional nonlinear least-squares fitting routine [3] we were only able to fit at most two terms in Eq. (1) at a given time; otherwise the parameters blow up.

As shown in Fig. 3, the shape of the OG waveform for the $1s_2$– $2p_2$ neon transition at 659.9 nm starts off being negative, but fairly quickly (within a microsecond) starts gaining in voltage, and then crosses over to the positive side (at around 3.45 µs) and subsequently diminishes to zero gradually. This is in contrast to the case for the $1s_4$–$2p_6$ neon transition at 630.2 nm (Ref. [5]) recorded at 10 mA current, which starts off being positive and then within about 5 µs crosses over to the negative side and then gradually decays to zero. The major difference between these two OG transitions is that the one reported in this paper does not involve an initial metastable state ($1s_2$), whereas the earlier one reported in Ref. [5] does involve a metastable state ($1s_4$). As seen from Table 1 and Fig. 4 below, the exponential rates b_2 and b_3 do not show significant variation over the range of currents studied; however, the effect is real, because as Fig. 3 clearly illustrates, the fitting is definitely improved using three terms in Eq. (1) (as compared to two terms). There is a small discrepancy between the experimental and fitted curves for two terms, whereas the fit using three terms is virtually perfect.

Table 1 below summarizes the instrumentation constant (τ), the amplitudes (a_j) and the exponential rates (b_j) extracted by the nonlinear least-squares fitting process using either two or three terms in Eq. (1) for a discharge current 10 mA, while Fig. 4 exhibits the variation of the exponential rates b_j ($j = 1$, 2,3) as a function of the current (2–19 mA).

Interestingly, as seen clearly in Fig. 4, the largest rate b_1 goes through a significant jump around 5 mA, a phenomenon that we noted earlier in Ref. [5]; it is an indication that a hitherto unexplained process is occurring in the discharge plasma and needs further exploration.

Table 2 above summarizes the numerical values of the exponential rates b_j ($j = 1$, 2,3) plotted in Fig. 4 which are averages of two separate data sets obtained for the 659.9 nm OG transition of neon recorded in the current range 2–19 mA.

Our Monte Carlo method implemented to perform least-squares fitting of the signal waveforms is more stable than the traditional nonlinear least-squares fitting algorithm, despite the fitting parameters being highly correlated. As a result, the fitting algorithm has the potential for wider application in the fitting of experimental signals that involve highly correlated basis functions.

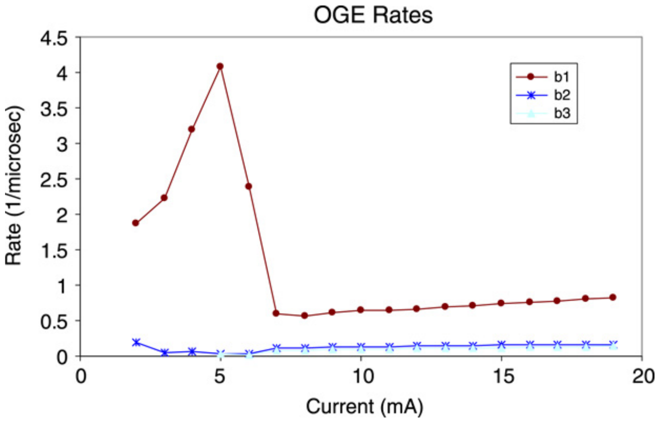

Fig. 4. Plot showing average rates b_j ($j = 1, 2, 3$) as a function of current (2–19 mA).

Table 2
Average decay rates b_j ($j = 1, 2, 3$) obtained from the nonlinear least-squares fitting of the observed OG waveforms corresponding to two data sets in the current range 2–19 mA.

Current (mA)	b_1 (μs^{-1})	b_2 (μs^{-1})	b_3 (μs^{-1})
2	1.874	0.196	
3	2.227	0.0446	
4	3.192	0.0609	
5	4.075	0.0373	0.0316
6	2.392	0.0249	0.0199
7	0.592	0.111	0.0870
8	0.570	0.116	0.0887
9	0.615	0.125	0.0934
10	0.648	0.133	0.0965
11	0.649	0.134	0.103
12	0.666	0.139	0.107
13	0.687	0.144	0.111
14	0.709	0.148	0.115
15	0.739	0.153	0.119
16	0.751	0.156	0.124
17	0.772	0.160	0.128
18	0.802	0.163	0.133
19	0.816	0.165	0.137

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