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Two-photon time-resolved optogalvanic signals of neon

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ABSTRACT

In this work, temporal evolution of two-photon laser optogalvanic signals of neon has been studied. Optogalvanic signals for four transitions from the metastable $2p^53s[3/2]_2$ state to $2p^54d'[3/2]_1$, $2p^54d'[3/2]_2$, $2p^54d'[5/2]_3$ and $2p^54d'[5/2]_2$ states were recorded over a range of discharge currents (3.4–9 mA). It was found that the shape of the optogalvanic signal was strongly dependent on the discharge current so that its peak shifted to shorter times and its amplitude increased with the discharge current. The decay rates of the 4d states, calculated from the optogalvanic signals, were found to increase linearly with the discharge current in the range of 6.2–9 mA. However, for the range of 3.4–5.4 mA, the decay rates were observed to slightly decrease with the discharge current.

1. Introduction

Illumination of a gas discharge with radiation at a wavelength corresponding to an atomic transition of a species in the discharge causes perturbation to the steady state population of two or more levels. This, in turn, causes a change in the electrical properties of the discharge. This process is known as optogalvanic effect [1] which has been found to be an excellent tool in the field of spectroscopy. Optogalvanic effect has shown high sensitivity in atomic and molecular spectroscopy [2,3] penning ionization spectroscopy [4], Doppler-free spectroscopy [5], plasma diagnostics [6], population inversion [7], and Rydberg-state spectroscopy [8]. It is also used as a valuable tool for the calibration of dye lasers [9,10].

Another significant aspect related to the optogalvanic effect is the study of discharge mechanism which helps a better understanding of atomic and collisional processes in plasma [11]. In this regard, many theoretical and analytical models, based on experimental observations, have been developed. These models characterize the response of discharge when steady state population distribution of the energy levels is perturbed by a radiation resonant to a transition between these levels. Ben-Amar et al. proposed a four-state phenomenological model to understand and describe the details of the optogalvanic signal [12]. They correlated the experimental results with those obtained from their proposed model taking into account relevant levels of the 3s and 3p manifold of neon. Doughty and Lawler presented a model based on perturbation theory to the key rate equations for neon discharges [13]. Another rate equation model to explain the optogalvanic effect in neon was formulated by Stewart et al. [14]. This work presented the dominant effects of electron collisional transfer in determining the sign and magnitude of the optogalvanic effect. In addition, Han et al. developed a simple theoretical model to understand the physics of the time-resolved optogalvanic signals [15,16]. Using this model, the decay rates of various transitions resulting from the excitation of neon's first metastable level was determined [17]. Piracha et al. have reported a study on the time-resolved optogalvanic spectra of neon and krypton using the Han et al. model [18]. They concluded that electron impact ionization was the dominant process in the characterization of the optogalvanic signal. Recently, Piracha et al. studied the time dependent optogalvanic signals induced by the $1s_4 - 2p_j$ laser excitations in neon DC plasma [19].

Much attention has been paid to the study of one-photon time-resolved optogalvanic signal including its behavior with the discharge current. Studies of two-photon time-resolved optogalvanic signals have rarely been reported [20,21] and, to the best of our knowledge, no study has yet been reported of the dependence of the signals on discharge current. In this work, we focus on the temporal evolution of two-photon optogalvanic transitions. Recently, eleven two-photon transitions originating from the $2p^53s[3/2]_2$, $2p^53s'[1/2]_0$, $2p^53s[3/2]_1$, and $2p^53s'[1/2]_1$ states to the $2p^54d$ configuration states have been investigated in the optogalvanic spectrum of neon in the visible region (570–626 nm) [22]. As examples, the two-photon transitions from the $2p^53s[3/2]_2$ metastable state were studied at different discharge currents. We used the theoretical model proposed by Ben-Amer et al. [12] to analyze the data and obtain parameters which determine decay rates of the upper state involved in two-photon transitions and the

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instrumental time constant. In addition, electron collisional rate parameters have been calculated by exploiting the linear relationship between decay rates and discharge current.

2. Experimental set-up

The experimental set-up was similar to that described in our earlier work [22]. The experimental arrangement consisted of a Nd:YAG laser (Quantel model YAG980, France) of 10 ns pulse width, pumped dye laser (Quantel TDL-90, France) and commercial Ag–Ne hollow cathode lamp. The dye laser produced 100–200 μJ/pulse with a linewidth of 0.08 cm^{-1} corresponding to 3 pm at 600 nm. A commercial Ag–Ne hollow cathode lamp ('Narva') along with a home-made adjustable dc power supply (200–600 V) was used to create discharge in neon. The discharge current was controlled by a current limiting load resistor (100 kΩ). A ballast resistor (22 kΩ) was used to read the discharge current. The laser beam illuminated the hollow plasma through the window on top of the lamp. The discharge current was adjusted between 6 and 9 mA. The laser was tuned to each selected resonant transition. Due to absorption of laser in the plasma channel, the voltage across the hollow cathode lamp varied for each laser shot. The voltage variation was coupled through a 0.01 μF capacitor and then fed to a digital oscilloscope (Picoscope ADC212, UK) and recorded as a function of time for each laser shot. Then, the voltage–time graphs were averaged over 30 shots for each current.

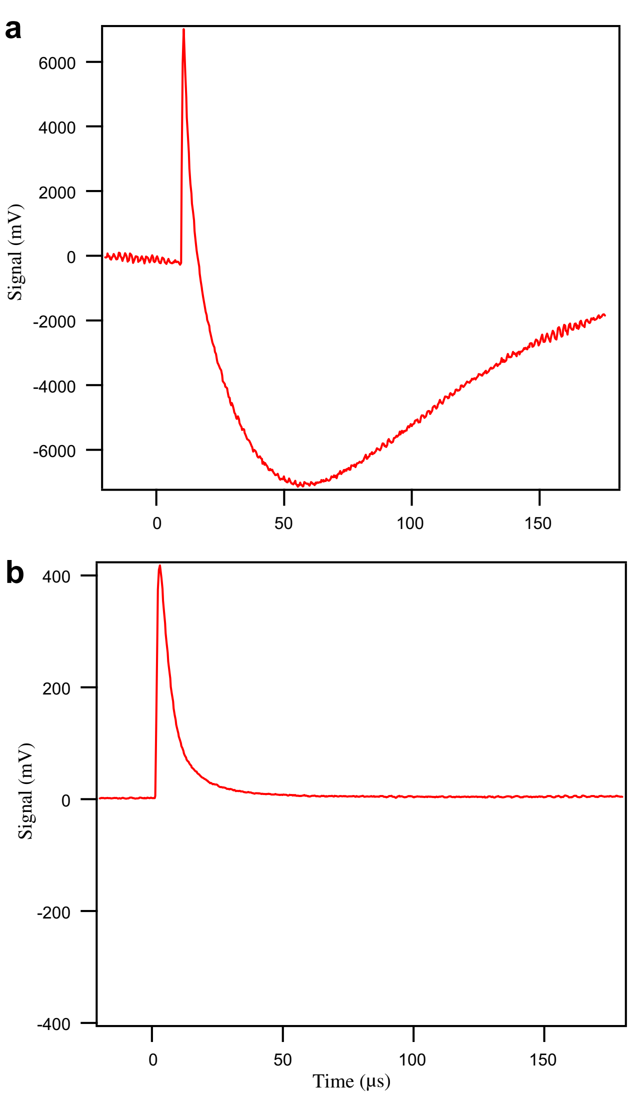

Fig. 1. Plots of the typical time evolution of an optogalvanic signal originating from a metastable state ($2p^53s[3/2]_2$); (a) one-photon transition, and (b) two-photon transition.

3. Result and discussion

The ground state electronic configuration of neon is $1s^22s^22p^6$ $^1S_0$. The excited states of Ne are best described through the j_cK-coupling scheme $[\{(l_1,s_1)j_c,l_2\}K,s_2]_J$ proposed by Racah [23]. In this coupling scheme, the orbital angular momentum l_2 of the excited electron couples with the total angular momentum, j_c, of the core to give the angular momentum, K, as $[j_c \pm l_2]$. The angular momentum, K, is then weakly coupled with spin angular momentum s_2 of the excited electron to give the total angular momentum, J, as $[K \pm s_2]$. Hence, the energy states are denoted by $nl[K]_J$. The $2p^53s$ configuration gives rise to four states designated as $2p^53s[3/2]_2$, $2p^53s[3/2]_1$, $2p^53s'[1/2]_0$, and $2p^53s'[1/2]_1$. Here, prime refers to the terms attached to the $3p^5(^2P_{1/2})$ parent ion level. Two of these levels, namely $2p^53s[3/2]_2$ and $2p^53s'[1/2]_0$, are metastable with radiative lifetimes of the order of seconds [24], while the other two levels, $2p^53s[3/2]_1$ and $2p^53s'[1/2]_1$, are short-lived with a radiative lifetime of 25 and 1.6 ns, respectively [25].

A theoretical model was proposed by Ben-Amar et al. to obtain information about the initial and final states involved in optogalvanic transitions [12]. In this model, it was assumed that the observed optogalvanic signal originating from the metastable state was the sum of the signals originating from the initial and final states. The following expression was derived from the rate equation approach to extract the physical dynamics of the time-resolved optogalvanic signal [12]:

$$S(t) = \beta\Delta n_0[a_2\exp(-t/T_2) - a_1\exp(-t/T_1)] - (a_2 - a_1)\exp(-t/\tau) \quad (1)$$

where, $S(t)$ is the amplitude of the optogalvanic signal and β is a constant related to the multiplication factor in the plasma. The coefficients a_1 and a_2 are related to the ionization rate of initial and final states, respectively. T_1 and T_2 are the relaxation times of the initial and final states and τ is the instrumental time constant. Finally, Δn_0 is the initial value population departure of the upper state.

Fig. 1 shows typical time evolution graphs for the one- and two-photon transition optogalvanic signals of Ne. As shown, the signal intensity of the one-photon transition is much higher than that of

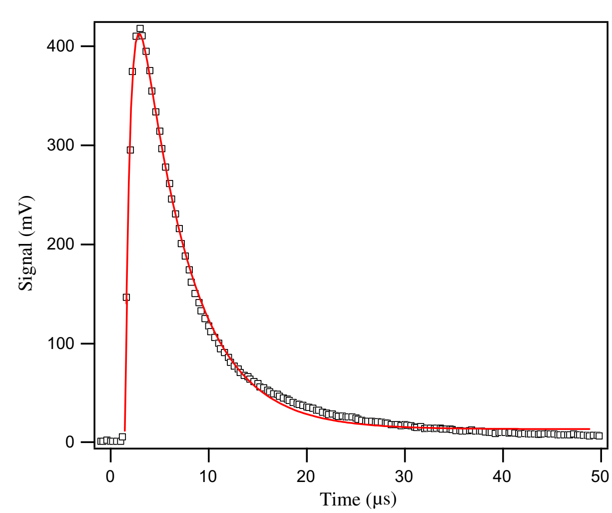

Fig. 2. Time-resolved optogalvanic signal of neon at 592.155 nm corresponding to $2p^53s[3/2]_2 \rightarrow 2p^54d'[3/2]_1$ transition. The solid line passing through the observed data points is the least square fit to Eq. (1).