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Temporal evolution of two-photon time-resolved optogalvanic signals of neon in the 600–630 nm region

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ABSTRACT

Time-resolved optogalvanic (OG) signals of six two-photon transitions of neon were studied in the 600–630 nm region to $2p^54d[5/2]_2$, $2p^54d[3/2]_2$, $2p^54d[3/2]_1$, $2p^54d[1/2]_1$ and $2p^55s'[1/2]_1$ states from the allowed states of the $2p^53s$ configuration ($2p^53s[3/2]_1$ and $2p^53s'[1/2]_0$ states). The OG signals were recorded over a range of discharge currents from 2 to 10 mA. The decay rates of the upper and lower states were obtained by fitting the waveforms with the Han et al.'s mathematical rate equation model considering the three states contributing to the signal. Based on the values of decay rates of the upper states, it was proposed that, after excitation to 5s and 4d states, neon atoms radiatively decay to the lumped relevant electronic states of the $2p^53p$ and $2p^54p$ configurations which have the main contribution in producing the OG signals. It was found that, the decay rates of the upper states (the lumped relevant electronic states of $2p^53p$ and $2p^54p$ configurations) increase linearly and slowly with the discharge current for all the transitions considered in this work. The effective decay rates of the upper states and their electron collisional ionization rate parameters were also obtained. This study showed that the dominant relaxation process in the de-population of the upper states is the lengthened radiative decay in plasma medium after laser excitation.

1. Introduction

OG spectroscopy is a very effective method for observing optical transitions in plasma media. The OG effect occurs due to the variation of impedance in a gas discharge when illuminated by a laser beam resonant to an atomic or molecular transition of the gas within the discharge [1]. In such a condition, the steady state population of the two states involved in the transition is perturbed. Consequently, the ionization balance changes and, if the laser operates in the pulse mode, a fluctuation occurs in the impedance of the discharge [2]. The OG signal is then detected by recording the change in the impedance of the discharge.

Much attention has been paid to the study of the OG spectra of inert gases via one and two-photon excitation. These spectra provide wavelength calibration for tunable dye laser-based experiments [3,4]. In addition, the study of time-resolved OG signals leads to understanding the discharge mechanisms as well as investigating the contribution of the electron collisional processes in the plasma [5]. One photon time-resolved OG signals of neon have been widely studied in the literature [2,6–13]. For example, Piracha et al. studied the one photon time-resolved OG signals of neon, all $1s_i$–$2p_j$ transitions (based on the Pachan notation [2]), with discharge current in the 610–680 nm wavelength region in order to understand the dominant physical processes responsible for the OG effect [6,7]. They found a linear relationship between the decay rates and discharge current which led to an estimate of the effective decay rates and the electron collisional rate parameters of the $1s_i$ states. Also, Mahmood et al. investigated the temporal evolution of the one-photon OG signal for the transition from $2p^53s\,[1/2]_2$ metastable state to the electronic states of $2p^53p$ configuration corresponding to the $\Delta J = \Delta K = 0, \pm 1$ transitions [2,8]. They investigated the contribution of the electron collision ionization as a physical process responsible for generating the OG signal. Also, they obtained an empirical relation which satisfactorily described the shift of the OG signal and the change in the population of the upper state as a function of discharge current [2]. Recently, Misra et al. have determined the decay rate of the $2p_2$ state of neon atoms by fitting the time-resolved optogalvanic waveforms of $1s_2 \rightarrow 2p_2$ transition at 659.9 nm through a nonlinear least-squares Monte Carlo approach [9].

Different mathematical models have been proposed for describing the temporal evolution of the OG signals [10–14]. These models describe the response of the discharge to a perturbation caused by laser illumination. Ben Amer et al. proposed a simple phenomenological model based on the electron multiplication factor in plasma in order to describe the OG signal and its time-dependent behavior

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[11]. Doughty and Lawler also presented a model based on perturbation theory to the rate equations for neon discharge [12]. In addition, a simple theoretical model was proposed by Han et al. for understanding the physics of experimentally observed time-resolved OG signals [15]. Unlike other models, this one clearly identifies and quantitatively characterizes the dominant physical processes contributing to the production of OG signal. Recently, this model has received much attention and has been used frequently by the researchers in the field of OG spectroscopy because it provides quantitative physical information about the states involved in the plasma.

Previously, the present authors reported eleven two-photon transitions of neon originating from the $2p^53s[3/2]_2$, $2p^53s'[1/2]_0$, $2p^53s'[1/2]_1$ and $2p^53s[1/2]_1$ states to the $2p^54d$ and $2p^55s$ configuration states in the 570–630 nm region [16]. Also, the time-resolved OG signals of four reported transitions from the metastable state $2p^53s[3/2]_2$ state to $2p^54d'[3/2]_1$, $2p^54d'[3/2]_2$, $2p^54d'[5/2]_3$ and $2p^54d'[5/2]_2$ states over a range of discharge current between 3.4 and 9 mA were studied using the Ben Amer's model [17]. It was found that the shape of the signal is strongly dependent on the discharge current so that its peak shifts to shorter times and its amplitude increases with the increase of discharge current. Considering only one exponential term, the effective decay rate and electron collisional rate parameter of the upper states were obtained. In addition, it was found that the decay rates of these states increase linearly with the discharge current in the range of 6.2–9 mA and slightly decrease in the range of 3.4–5.4 mA.

In the present study, there is a focus on the temporal evolution of the six two-photon OG signals of neon in the range of 600–630 nm wavelength region. The behavior of the signals with discharge current, which is different from that of the one-photon signals studied in the literature, was investigated based on the Han et al.'s model. To the best knowledge of the authors, this model has not been used for the analysis of two-photon transitions. The model was used for obtaining the effective decay rate and electron collisional rate parameter of the upper and lower states contributing in the OG signal.

2. Experimental setup

The experimental set-up to study the behavior of the OG signals is similar to the one described in our previous work [16] (Fig. 1). An Nd:YAG laser (Quantel model YAG980, France) of 10 ns pulse duration was used for pumping a dye laser (Quantel TDL-90, France). The linewidth of the dye laser is about 0.08 cm^{-1} corresponding to 0.0030 nm at 600 nm. Rhodamine 640 dye was used for creating

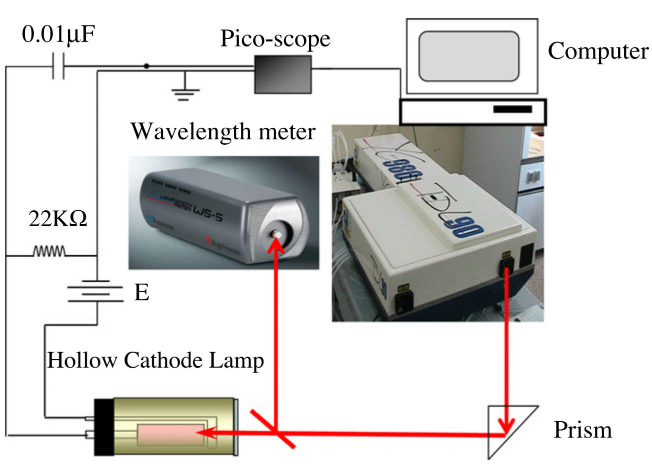

Fig. 1. Schematic of the experimental arrangement for the laser optogalvanic spectroscopy.

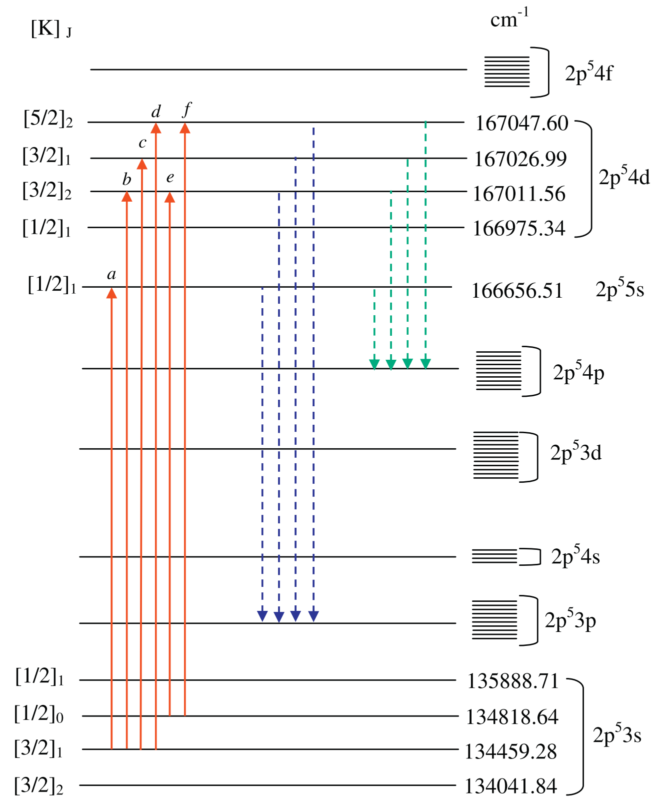

Fig. 2. Partial energy level diagram for neon transitions. Red arrows show the excitations at (*a*) 620.999, (*b*) 614.910, (*c*) 613.934, (*d*) 613.547, (*e*) 621.083 and (*f*) 620.387 nm. Blue- and green-dashed arrows show the radiative decay from the states of $2p^54d$ configuration to the states of $2p^53p$ and $2p^54p$ configurations, respectively. All the states of $2p^53p$ and $2p^54p$ configurations were separately lumped into a common state. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

the laser light between 605 and 626 nm. In addition to the previous experimental setup, a wavelength meter (WS5, Angstrom) was used for the calibration of the dye laser. A part of the laser light, separated by a splitter, was directed toward the wavelength meter. A commercial Ag–Ne hollow cathode lamp (Narva, Germany) along with a home-made adjustable dc power supply (200–600 V) was used for creating discharge in neon. The discharge current was controlled by a current limiting load resistor (100 kΩ). A ballast resistor (22 kΩ) was used for reading the discharge current. The laser beam illuminated the hollow plasma through the window on the top of the lamp. The discharge current was set to fixed values between 1 and 10 mA for each experiment. The wavelength was tuned in resonance with those atomic transitions presented in Fig. 2. The voltage across the discharge was coupled through a 0.01 μF capacitor; then it was fed to a digital oscilloscope (Picoscope ADC212, UK). The variation in the voltage, caused by the laser pulse, was recorded as a function of time. The voltage–time graph was averaged over 30 laser shots for each wavelength.

3. Results 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 [8]. In this coupling scheme, l_2 is the orbital angular momentum of the excited electron and j_c is the total angular momentum of the core. Coupling of l_2 and j_c gives K. Under this scheme, the first excited state for neon $2p^53s$ gives rise to the four levels; namely $2p^53s[3/2]_2$, $2p^53s'[1/2]_0$, $2p^53s'[1/2]_1$ and $2p^53s[1/2]_1$. Here, prime refers to