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Chopped CW laser-induced optogalvanic effect in a neon hollow cathode discharge

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Abstract. A detailed model for the optogalvanic effect in a neon hollow cathode discharge irradiated by a chopped CW dye laser is presented. A rate equation formalism is used to calculate the evolution of the first and second electronic configuration populations coupled by the laser and of the electric charges number density. Processes as ambipolar-like electrons loss, electronic collisional coupling of level populations and electron emission by the cathode due to VUV radiation from the $1s_2$ resonant level are taken into account and further discussed.

The transients and steady-state magnitude of the optogalvanic signal are calculated, compared with experimental data and related to population changes. We predict sign changes of the optogalvanic signal when the laser is tuned over transitions originating from the resonant level with respect to transitions involving the metastables states. The optogalvanic signal is shown to be basically determined by the laser-induced variations of the excited-state populations while changes in the electron temperature, due to laser energy transfer by collisions between electrons and excited atoms, play a negligible role.

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The development of new spectroscopic techniques has allowed to increase the knowledge about the structure of atoms and molecules. Particularly with the arrival of lasers, completely new techniques were developed and some methods were reconsidered such as the optogalvanic spectroscopy.

The detection of the optogalvanic (OG) effect is based upon the current changes that occur when a discharge is illuminated by radiation which is resonant with one transition of the atoms and molecules. Therefore, in opposition to the difficulties for detecting small intensity variations by radiation absorption or by fluorescence superposed to an important optical background, the OG spectroscopy presents less measurement problems, even in very low-concentration samples.

The OG effect presents a wide range of applications, besides spectroscopy: detection of small concentrations, studies of electric discharges in gases, identifications of excited states, isotopical analysis, wavelength calibration and lasers stabilization. Its applications and the study of the phenomenon itself have recently given rise to conferences [1, 2] and review papers [3].

A quantitative description of the OG effect should necessarily be based on a discharge model, which will be different in the positive column or in the cathodic region. It is generally supported by rate equation formalisms which include the de-excitation and excitation rate coefficients of the discharge species, and therefore require the knowledge of all the processes and cross sections as well as the energy distribution of electrons and atoms.

When the plasma is disturbed by radiation which is resonant with one of the species therein, the rates of the processes included in the discharge model will be affected by a variation in the populations of certain levels and in the number of ion–electron pairs, that at the same time has an effect on all the populations, or by a variation of the electrons and gas temperature. The generation of the OG signal can be explained by either mechanism, that is going to be dominant according to the kind of discharge and to the excited atomic species. It is clear that a general theoretical description should include a great number of processes that vary according to the type of discharge and would give rise to a high-complexity model. The impossibility of describing all the phenomena named above gives rise to simplifications like the description of a particular discharge, or the inclusion of just only the levels that will be related by the laser radiation, or the introduction of empirical parameters, such as decay times that would represent one or several relaxation phenomena, derived from the fitting of the OG signal by a sum of exponentially decaying functions [4, 5].

The positive column discharges can be characterized by a uniform electric field, a Maxwellian electron distribution, single or multi-step ionization phenomena by atom–electron collisions and losses of charges by ambipolar diffusion toward the walls of the discharge tube [6, 7]. Several authors analyze the variation of the dynamic impedance of a neon discharge by irradiation with a CW laser using different approaches and generally predict the magnitude and the sign of the OG signal for a given excited transition and its sign changes upon varying the initial steady-state current [7–10]. The used approximations consist of including all or some of the $1s_i$ (Paschen notation) levels, the laser perturbed or all the $2p_j$ levels and different excitation and de-excitation processes by collisions with atoms or electrons.

The description of the OG signal production in neon in the cathodic region must necessarily be based on a different discharge model than the former one due to the non-uniformity of the electric field, which is very intense in the cathode fall and reduces drastically a few millimeters far from the cathode. The resulting electronic distribution is not Maxwellian because of the existence of high- and low-energy electrons due to ionization processes. In particular, in opposition to the positive column, in hollow cathode discharges, a model that describes the OG signal that originates within the cathode will not be able to include an ambipolar radial diffusion term toward the walls since the field is not zero there. A simplified model of five levels, with a Maxwellian electrons distribution and ambipolar diffusion that predicts the OG intensity upon exciting some neon transitions and its sign change with the current was published by van Veldhuizen et al. [11]. An alternative model of four levels for steady state predicts a sign change in the OG signal when the radiation excites atoms from metastable or from resonant states and concludes that the magnitude of the signal is related to the population of the metastable states and the pumping rate [12]. Other works carried out at high current or with cathodes of specific materials show that the steady-state OG signal is related to a change of the electrons temperature [13–15] due to numerous inelastic and superelastic collisions of electrons with excited atoms of the sputtered material when the laser is tuned on one of its transitions.

In conclusion, we can assure that there does not exist a detailed model for a hollow cathode negative glow discharge in neon that describes the dynamics of the OG signal and its sign, which is different when the excitation starts from a metastable or a resonant level, and allows to predict transients that appear on the switch-on and -off of the laser that illuminates the discharge [5, 16, 17]. Therefore, in this work, a model based on rate equations is outlined which permits to describe this type of discharge and the observations carried out upon disturbing the plasma when different transitions of its atoms are excited by a CW dye laser. The phenomena related to the terms included in the equations are discussed. Furthermore, some approximations are applied that, yet simplifying the model, allow to predict the transients that appear in the OG signal, when the CW dye laser is switched-on and -off, and the magnitude and sign of the signal according to the laser-excited transition.

1 Theory

The magnitude and time behavior of the OG signal will depend on the participation rate of the externally perturbed electronic level populations of a given atomic species in the discharge. Furthermore, the influence of the change produced on other levels population by radiative coupling or collisional interaction with other plasma species should be taken into account. We have shown, even using a very simple model [18] where only the populations of the laser-coupled levels vary, that the signal is the result not only of the raising of populations to levels nearer ionization but also of the change in the first configuration population.

Therefore, in order to predict correctly the OG signal under a given perturbation it is necessary to make a detailed dynamic description of the populations contributions to the current considering all the change mechanisms involved. The present model will evaluate the evolution of the populations and the current by means of rate equations for the negative glow region in a neon hollow cathode discharge.

The lowest-excited electronic configuration of neon consists of four $1s_i$ energy levels ($2 \leq i \leq 5$). Two of these states, $1s_3$ and $1s_5$, are metastable. The $1s_4$ state is a quasi-metastable state with a slight triplet–singlet mixing. The $1s_2$ state is radiatively coupled with ground state ($\lambda = 73.6$ nm, $\tau = 1.5$ ns). The next-excited configuration consists of the $2p_j$ levels ($1 \leq j \leq 10$) radiatively coupled to the $1s_i$ levels. These two configurations were connected by the laser radiation to observe the OG signal.

The discharge will be described as a neutral plasma characterized by an electron energy distribution which is the sum of two Maxwellian distributions corresponding to temperature T_{h} and $T_\ell (T_{\mathrm{h}} > T_\ell)$ [19] and number densities n_{h} and n_ℓ ($n_\ell/n_{\mathrm{h}} \sim 10$–$100$) [6] in a region with a constant small value for the electric field (E). The fast electrons, with a mean velocity $\sqrt{3kT_{\mathrm{h}}/m}$, are those that have been emitted from the cathode, accelerated by the cathode fall potential and had lost energy through several inelastic collisions. The slow electrons distribution (T_ℓ) consists of electrons ejected by atomic ionization.

As n_{h} is smaller than n_ℓ, the discharge current will be evaluated by means of the following equation:

$$I = e\pi r^2 n_\ell(n_{\mathrm{i}}, n_{\mathrm{h}}, T_{\mathrm{h}}, T_\ell)\, v_{\mathrm{d}}(E/p) \,,$$

where e is the electron charge, r the discharge radius, p the gas pressure and n_i the atomic level populations. As will be further discussed, the perturbation of slow electrons density caused by the electronic temperature variation is small compared to the changes which take place due to the laser-induced population variations. As long as the voltage drop on the lamp can be considered constant, which corresponds to a ballast resistance of few $k\Omega$, the electric field variation associated with the change of charges density during the OG effect is negligible and the drift velocity v_{d} change related to it is unimportant [20]. In this case, the OG signal can be considered as generated by the evolution of the electronic number density n_ℓ associated only to changes of the n_{i} electronic level populations and n_{h} electronic number densities.

The perturbed discharge will be described by a rate equation system for each of the $1s_i$ and $2p_j$ populations, and for n_ℓ similar to our previous model [18], adding equations for the fast electron density variation and for the atomic and molecular ions densities. The main processes involved in this model are shown on the neon energy level schemes published in [18], where all the rates are now taken from [21]–[44].

The equation for the slow electrons density to be considered consists of terms of charges losses by three body [21], radiative [22] and dissociative [23] recombination and of creation of charges that will take into account atomic ionizations from different electronic levels by collisions with electrons [24–26] and from $1s_i$ by collisions with $1s_j$ atoms [27–30]. Additionally, we consider a term associated with charges diffusion proportional to n_ℓ through a factor D and another one with secondary electron emission at the cathode.

The first additional term, which describes the simultaneous migration of an electron and an ion inside a negative-charged contour, has been proposed and discussed [18,31] in place of the radial ambipolar diffusion which cannot be applied in a hollow cathode. We determined experimentally the coefficient D, in a commercial lamp similar to the one used in [5], from the decay time of the OG signal when a short light pulse excites the $2p$–$3s$ transition without perturbing the metastable level populations. The D value obtained by this method was $2.1 \times 10^5\ \mathrm{s}^{-1}$. Our works show that only the inclusion of this process allows to fit the theoretical to the experimental steady-state recovery rate of the $1s_i - 2p_j$ OG signals obtained with a pulsed laser.

The second additional term takes into account the secondary electron emission variation with respect to the unperturbed discharge and describes its effect on the negative glow region. Sturges et al. [32] have shown that the secondary emission by metastable bombardment of the cathode is not essential in hollow cathode discharges. Additionally, the metastable diffusion time is around 100 μs [33], which is much longer than the transients of the OG signal under CW or pulsed irradiation. Then and according to von Engel [34], we assume that the value of the secondary emission coefficient in hollow cathode discharges is determined mainly by VUV photons, emitted principally by the decay of the $1s_2$ population to ground state, that induce photoelectric effect at the cathode. As the electrons emitted by the cathode are accelerated in the cathode fall and lose energy through several ionizing collisions with gas atoms setting up the electron distribution proposed for the negative glow region, so a variation of the $1s_2$ population will induce changes in n_ℓ and n_h number densities.

In the slow electrons density equation, the term associated with secondary electron emission is written proportionally to the change of the $1s_2$ population through a factor W, that includes the quantum efficiency of the cathode [35], the number of possible ionizations produced by each electron accelerated through the cathode fall and the Einstein coefficient of spontaneous emission corrected by radiation trapping [9].

The equation for the variation of n_h contains a diffusion term, like the n_ℓ electrons equation, and a creation one due to secondary emission from the cathode proportional to the change of the $1s_2$ population through a factor W'. The ratio of the terms associated with the photoelectric effect in the n_ℓ and n_h equations (W/W') is imposed equal to the quotient n_ℓ/n_h in absence of resonant laser excitation since the photoelectrons are also part of the unperturbed electron distribution.

The rate equation for the resonant level ($1s_2$) is written taking into account the contribution of electronic excitations from ground state [36] and the fluorescence from higher levels [37] and is balanced by excitation to higher states [38] and ionization by electrons and ionization from $1s_i$ by collisions with $1s_j$ atoms. The $1s_i$ collisional population transfer to $1s_j$ by atoms [39] and electrons [40,41] and radiative decay to ground state, corrected by radiation trapping [9], is now added with respect to the former model. Similar equations are proposed for the other three $1s_i$ levels where the term describing the VUV fluorescence is replaced by metastable diffusion [33].

The dynamics of the $2p_j$ level populations are obtained considering excitation from ground state [42] and from $1s_i$ by collisions with electrons, deexcitation by fluorescence to $1s_i$ and ionization by electrons. The $2p_j$ level population exchanges by collisions with electrons are included, the rates being calculated based upon the classical impulse approximation [26]. These rates are around 100 times faster than the collisional coupling by atoms [43] for typical slow electrons [18] and gas [5] temperature values. They are also higher than the spontaneous emission rates; consequently, thermodynamic equilibrium for the unperturbed discharge among the $2p_j$ level populations will be considered. When a $2p_j$ level is externally pumped, the coupling process redistributes the external perturbation among all the other neighboring, and allows fluorescent relaxation via multiple decays to the $1s_i$ levels. If coupling rates are now compared with the pumping rate for a laser power of some mW, only the populations of $2p_2 - 2p_5$, $2p_6$ and $2p_7$, $2p_8$ and $2p_9$ can be considered in thermodynamic equilibrium with the slow electrons during laser irradiation. This approximation allows reducing the description of the evolution of the neon second-excited electronic configuration, composed of ten levels, to five rate equations.

We will also include a balance equation which takes into account the molecular ion Ne_2^+ formation by collisions of an atomic ion Ne^+ with atoms in the ground state [44] and its destruction by dissociative recombination. The equation for the atomic ions is similar to the slow-electron equations which includes Ne_2^+ formation instead of dissociative recombination.

A similar model with the named approximations gave a good quantitative agreement between the calculated and experimental transient OG signal, obtained in our laboratory when two different transitions were excited by a nanosecond-pulsed laser [45].

2 Discussion of results

We applied our model for a commercial hollow cathode lamp as used by Uchitomi et al. [5] and Smyth et al. [16], with a current about 10 mA and neon pressure of 5 Torr, where the measured gas temperature was about 1500 K [5]. Assuming thermodynamic equilibrium among all the $2p_j$ levels, we determined spectroscopically, in a similar lamp, that the slow electrons temperature is around 4000 K. The equations are numerically solved in absence of laser radiation with the charge neutrality condition varying the values of T_h and n_h with the typical range for a glow discharge. Typical values for neon hollow cathode discharges are obtained for $1s_i^0(\sim 8\times 10^{11}\,\mathrm{cm}^{-3})$ and $2p_j^0(\sim 6\times 10^{8}\,\mathrm{cm}^{-3})$ populations, for the $n_\ell^0(\sim 6\times 10^{11}\,\mathrm{cm}^{-3})$ electrons densities and n_ℓ^0/n_h^0 ratio $(=26)$ with $T_h = 47000\,\mathrm{K}$ and $n_h^0 = 2.3\times 10^{10}\,\mathrm{cm}^{-3}$ [6,12,32,46].

These values are the initial conditions for the solution of the equations when the discharge is illuminated by a chopped CW laser, keeping constant the electronic temperatures. The results of the relative electron density variation with respect to equilibrium $((n_\ell - n_\ell^0)/n_\ell^0)$, when the laser is tuned on the $1s_5$-$2p_2$ and $1s_2 - 2p_1$ transitions, are presented in Fig. 1 and Fig. 2, respectively, where the laser intensity within the absorption line width is 20 mW/cm^2. It should be observed that the resultant change of steady-state current is comparable to the empirical results from Uchitomi et al. [5] under similar laser excitation. The quantitative fitting of the theory to the experiment could be improved if the experimental conditions for the OG signal measurement were known in detail. It should be remarked that the sign of the calculated and experimental OG signals is the same for both transitions. The sign inversion for excitation of one transition with respect to the other cannot be explained if only the laser-induced $2p_j$ level over populations are taken into account. In the following paragraphs we show that the current variations are related to the changes in the $1s_i$ populations determined by fluorescence from $2p_j$, collisional coupling among $1s_i$ and $1s_2$ radiative decay.

Actually, in the $1s_5$–$2p_2$ case, part of the $1s_5$ population is transferred to the other $1s_i$ (Fig. 3) through excitation to $2p_j$. Particularly, the losses by fluorescence from $1s_2$ increase and, as a consequence, the resulting $1s_i$ total population decreases. On a large time scale, the hole in the $1s_5$ population is transferred to the other $1s_i$ by collisional coupling so that the individual populations reach values below the initial conditions before irradiation. Therefore, the steady-state current under laser excitation will diminish accentuated by the decrease of the photoelectrons quantity.

In the $1s_2$–$2p_1$ case the laser produces a decrease of the $1s_i$ populations losses through fluorescence from $1s_2$ to the ground state and, at the same time, the metastable level populations increase through fluorescence from $2p_j$ but the changes are attenuated by collisional coupling. In a large time scale, this effect, added to the $1s_i$ excitation from ground state, makes the current to increase due to ionization of a larger number of $1s_i$ atoms, in spite of the photoelectrons number decrease.

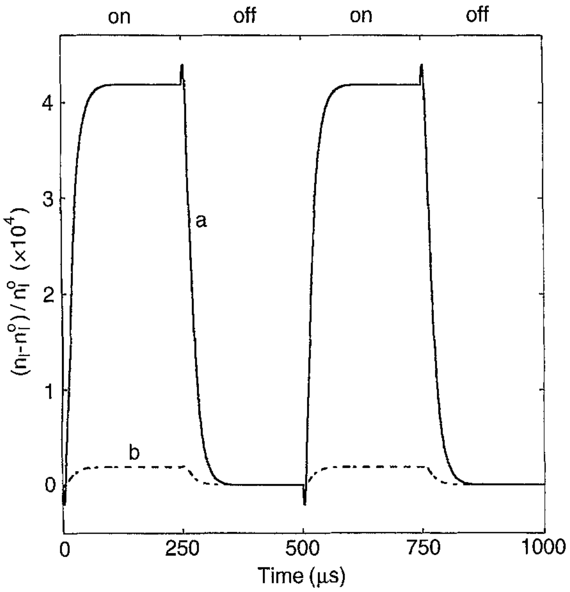

Fig. 2. Relative electron density variation for excitation on the $1s_2$–$2p_1$ transition vs time for **a** $I_{\mathrm{laser}} = 20\,\mathrm{mW/cm^2}$ (solid line) and **b** 10 K slow electron temperature increase (dotted line)

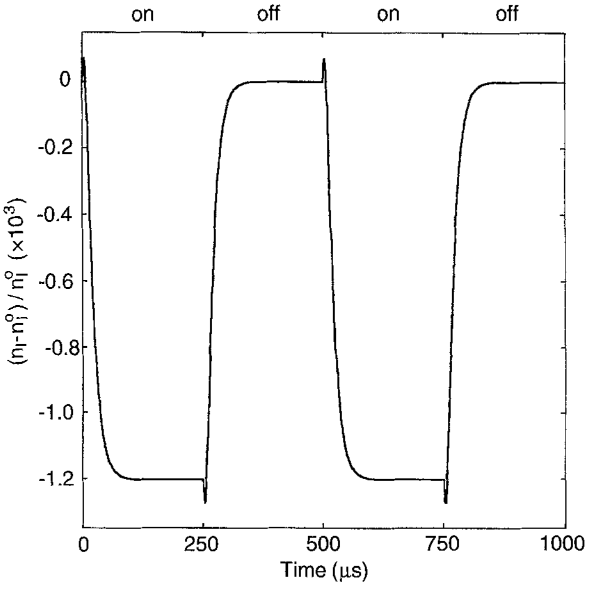

Fig. 1. Relative electron density variation for excitation on the $1s_5$–$2p_2$ transition vs time at $I_{\mathrm{laser}} = 20\,\mathrm{mW/cm^2}$

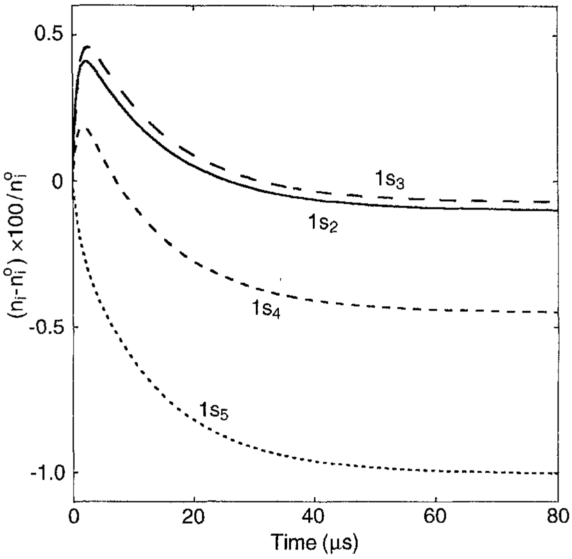

Fig. 3. Relative $1s_i$ population densities variation for excitation on the $1s_5$–$2p_2$ transition vs time

Moreover, the model describes the peaks that appear when the laser, tuned at the $1s_5$–$2p_2$ transition, is switched-on and -off (Fig. 1) as observed by Smyth et al. [16], Uchitomi et al. [5] and Labat et al. [17]. Figure 3 shows that during the laser switch-on time the $1s_2$, $1s_3$ and $1s_4$ level overpopulations through $2p_j$ allows for an increase of the electrons density due to ionization and photoelectric effect, which is more important than the decrease due to $1s_5$. During the switch-off time, the $1s_2$, $1s_3$ and $1s_4$ level populations fall off due to the loss of the fluorescence contribution from overpopulated $2p_j$ levels and the current decreases before recovering its initial value.

We also prove by means of this model that the form of these peaks depend on the ratio of the $1s_5$ level diffusion and collisional coupling relaxation time (~ 100 μs) to the laser switch-on and -off time. Figure 1 shows the calculated OG signal with a switch-on and -off time of 2 μs. No changes are observed for shorter times; for 20 μs the peaks broaden and decrease in amplitude (Fig. 4).

We have already affirmed that laser-induced electronic temperature variations do not contribute significantly to the OG signal. These are due to changes in the populations that modify the energy transfer between excited atoms and electrons. In Fig. 2, the OG signal obtained with our model is shown, under the assumption that the whole laser energy is totally transferred to the electrons and produces a T_1 temperature increase of 10 K, similar to the value measured by Drèze et al. [13] and calculated by Keller et al. [15] and higher than the value calculated by Maeda et al. [14] for hollow cathode discharges in noble gases irradiated by a CW chopped laser. It should be noted that this temperature change does not allow to reproduce the experimental signal; according to our calculations an increase of around 500 K would be necessary in order to reproduce the magnitude of the OG signal.

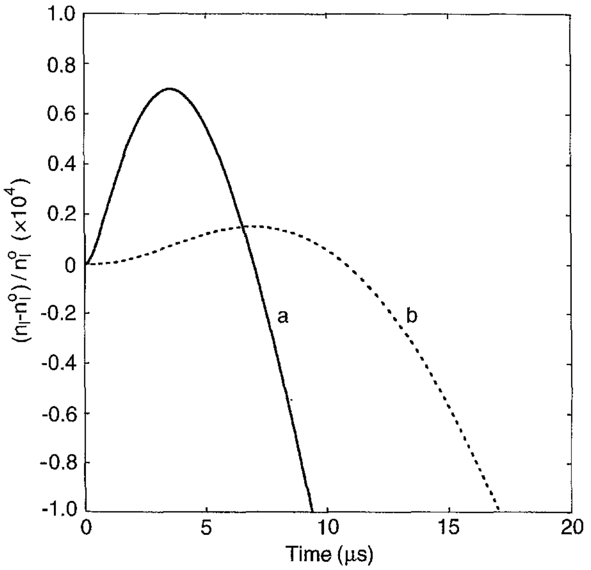

Fig. 4. Relative electron density variation for excitation on the $1s_2$–$2p_1$ transition vs time for **a** 2 μs laser switch on time (solid line) and **b** 20 μs switch on time (dotted line)

3 Conclusions

A detailed model has been developed that qualitatively describes the OG signals found when hollow cathode discharges in noble gases are illuminated by a chopped CW laser. It predicts the peaks which appear during the laser switch-on and -off time and the dependence of their forms, which are associated with atomic parameters, on the rise time of the laser radiation. We also prove that the OG signal, like in the experiment, inverts its sign when the laser is tuned to transitions starting from resonant levels with respect to transitions starting from metastable levels.

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