An absorption spectroscopy technique for high voltage pulsed discharge afterglow plasma investigation

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An absorption spectroscopy technique with temporal resolution using a conventional spectral source operating in pulsed mode has been used to determine the temporal distribution of neon metastable atoms density in the afterglow of a high voltage pulsed hollow cathode discharge plasma. The measured values for Neon 2^3P_2 state density in the 40 - 120 μs temporal afterglow range are found to lay from $1.2 \times 10^{12} \pm 20\%$ cm^{-3} to $3 \times 10^{10} \pm 20\%$ cm^{-3} .

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1. Introduction

The experimental data reported until now on the determination of the metastable atoms density in different configurations of pulsed discharges have been obtained using sophisticated techniques like laser absorption or laser induced fluorescence [1-3]. A hollow cathode lamp working in the high voltage regime [4], has been used as an intense optical source for spectral analytical measurements only in the investigation of dc and RF plasma [5, 6].

In this paper, through the use of a conventional pulsed spectral source and an original experimental supply and detection system we determined the temporal distribution density of Ne metastable atoms in a hollow cathode pulsed discharge afterglow plasma. This temporal distribution, an indicator of generation and loss processes of these long-lived radicals in reactive plasmas, may be very useful in analytical investigation with systems like GDOES and GDMS [7-10], in which the energy transfers of the various active species present in the plasma, electrons, ions and neutrals are central for the excitation and ionization of the analyte species.

2. Experimental set-up

The intense pulsed spectral source, which we will call in this paper, spectral lamp, consists of a glass chamber containing a titanium cylindrical hollow cathode (15 mm length and 3 mm diameter) and a stainless steel wire anode. The neon gas pressure is 1-5 torr and peak current pulses in the range of 30 A with duration of 120 ns (at half-width) and repetition rate of 30 Hz are applied. This pulsed high current density provides a large amount of excited neon atoms and ions with intense emission lines.

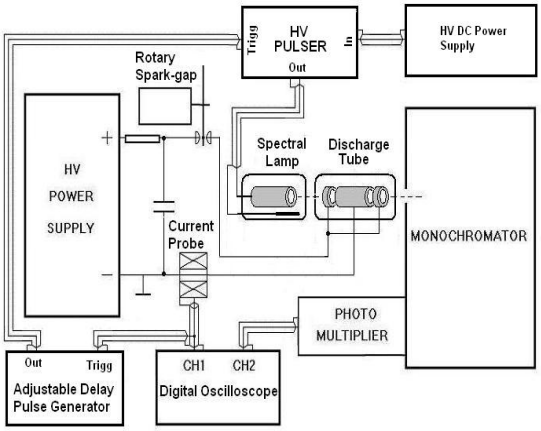


Fig. 1. Experimental set-up for temporal resolution optical absorption measurements.

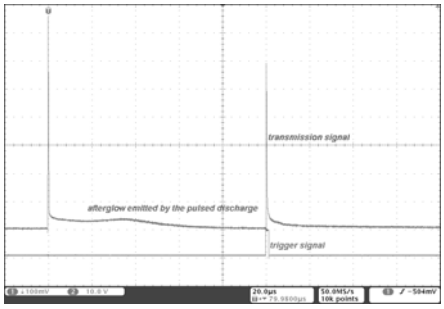
The absorbing species of interest in our present work are the neon metastable atoms existing in the afterglow plasma produced by a pulsed high voltage hollow cathode discharge. The discharge tube consists in a cylindrical hollow cathode of Titanium (20 mm length and 4 mm diameter) and two stainless steel anode rings (4 mm diameter) placed symmetrically at the two ends of the cathode and is operated at a pressure of 5 torr neon, and 60A peak current, 5 kV voltages applied by using a rotary spark-gap [11]. High current pulses of short duration in the hollow cathode discharge tube were obtained by repetitively, frequency 30 Hz, discharging a storage capacitor C through a rotary spark gap with a commutation time below 10 ns, the electrical set up being previously reported [12] for Neon and Argon working gas.

The spectral lamp is optically aligned with the hollow cathode pulsed discharge tube (source of absorption

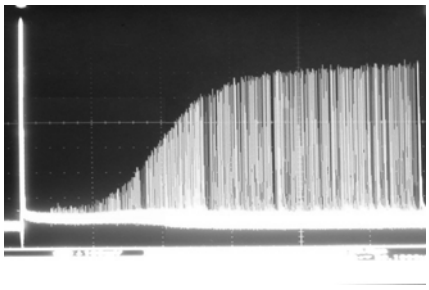
species i.e. neon metastable atoms). The light emitted by the spectral lamp and transmitted through the discharge tube is filtered with a 1 m Jarrell-Ash grating (1200 lines mm^{-1}) monochromator, detected with an EMI photomultiplier tube and recorded with a Tektronix oscilloscope.

A specific home-made electronic arrangement was used to apply on the spectral lamp high voltage pulses with variable temporal delay related to the hollow cathode discharge current pulse, thus allowing the detection of the transmitted light at various times in the temporal afterglow plasma of the high voltage discharge. As it can be seen in the Figure 1 the spectral lamp was triggered using the signals collected by a current probe from the hollow cathode discharge tube circuit and delayed by an adjustable pulse generator from the discharge tube current pulse [13].

In the oscilloscope image presented in Fig. 2 it can be seen the trigger signal from the adjustable pulse generator temporary delayed from the current pulse of the discharge tube, and the light signal from the photomultiplier filtered by the Jarrell Ash spectrometer, at 614.3 nm (corresponding to the transition $^3P_2 - 2p_6$, $^3P_2$ being the metastable level at 16.62 eV), which consists of the temporal profile of the emission light of the pulse hollow cathode discharge with the long afterglow and the transmission light of the spectral lamp positioned by the trigger signal at various times in the afterglow plasma.



(a)



(b)

Fig. 2. (a) The temporal profile of the 614.3nm line with the long afterglow emitted by the pulsed discharge, the delayed trigger signal (which starts the spectral lamp emission) and the transmission signal; (b) the photo of the oscilloscope screen taken with long exposure time while the transmission sweeps the afterglow.

Varying the delay of trigger so that it sweeps the entire period of the afterglow, the transmission light peak describes a transmission profile as it is shown in the photo of the oscilloscope screen (Fig. 3 (b)) taken with long exposure time.

3. Results and discussion

The temporal evolution of the concentration of neon metastable atoms in the afterglow of a high voltage pulsed discharge is achieved from the transmission profile obtained by the procedure presented above. The measured data are than processed with the well known absorption spectroscopy relations obtaining the neon metastable atoms concentrations at various measured times in the afterglow.

The profile of the spectral lines emitted by the spectral lamp source is Doppler type and was determined by measuring the gas temperature of the pulsed plasma (by the method reported in [14]) from the shock waves frequency produced during the breakdown of the high voltage pulsed discharge.

For the *Spectral Lamp* under the experimental conditions of 5 torr neon pressure and 30 A peak current, the shock waves obtained during the discharge current period is of about 190 kHz. The gas temperature was calculated using the Rayleigh formula for the sound velocity: $\upsilon_S = 2\pi\nu_s / k_s = (\gamma k T / m)^{1/2}$ where ν_S and k_s are the frequency and the corresponding wave number of the sound, $\gamma = c_p / c_v$ is the ratio of specific heats, k stands for the Boltzmann constant, T is the gas temperature, and m is the mass of the gas atom and was found to be of about 11 800 K.

In absorption spectroscopy, the measured relative absorption (in percents) is given by the formula $A = 1 - I_t / I_0$, where I_0 is the intensity of the emitted light of the lamp and I_t the transmitted light, which is obtained by subtracting the emitted light of the plasma from the light emitted simultaneously by the plasma and the lamp ($I_t = I_{plasma + lamp} - I_{plasma}$).

According to Mitchell and Zemanski [15] the absorption is given by

$$A = \sum_{m=1}^{\infty} \frac{(-1)^{m+1}(k_0 L)^m}{m!\sqrt{1+m\alpha^2}} = \frac{k_0 L}{\sqrt{1+\alpha^2}} - \frac{(k_0 L)^2}{2!\sqrt{1+2\alpha^2}} + \ldots \quad (1)$$

where k_0 is the maximum absorption coefficient of the line, L the absorption length (2 cm) and

$$\alpha = \frac{Doppler\ lamp\ line\ broadening}{Doppler\ species\ line\ broadening} \quad (2)$$

with the Doppler line broadening given by the formula $\Delta\vartheta_D(cm^{-1}) = \frac{7.16\times10^{-7}}{\lambda(cm)}\sqrt{\frac{T}{M}}$. Thus, the formula (2) becomes $\alpha = (T_l / T_s)^{1/2}$ with T_l standing for the gas temperature of the spectral lamp and T_s for the absorbing species temperature.

For the *Discharge Tube* the frequency of the shock waves in the experimental conditions 5 torr neon pressure, 60A peak current at 5 kV applied voltage is of about 78 kHz and can be considered almost constant during the afterglow (≅ 120 μs) of a pulsed discharge throughout the plasma volume. Thus the calculated gas temperature for the afterglow plasma is T_s = 2000K.

The neon metastable density *N* (atoms/cm^3) has been computed with the Ladenburg's formula for k_0,

$$k_0 = \frac{2}{\Delta\vartheta_D}\sqrt{\frac{\ln 2}{\pi}}\frac{\pi e^2}{mc}N f \qquad (3)$$

where $\Delta\vartheta_D$ is the Doppler species line broadening, *e* and *m* are the electron charge and mass, *c* is the light velocity and *f* is the oscillator strength of the transition.

We determined the $^3P_2$ level density profile along the afterglow using the absorptions measured on the transition ($^3P_2 - 2p_6$) at 614.3 nm with f= 0.122 [16]. The Doppler line broadening of species at 614.3 nm wavelength is $\Delta\vartheta_D$ = 0.1166 cm^{-1} calculated for T_s = 2000 K. The experimental transmission and absorption measurements on the transition ($^3P_2 - 2p_6$) at 614.3 nm and the corresponding calculated $^3P_2$ level concentration data starting from 50 μs to 120 μs in the temporal afterglow range of the pulsed hollow cathode discharge are presented in the Table 1 and plotted in Fig. 3.

Temporal position in afterglow (μs)	Measured Transmission (%)	Measured Absorption (%)	Atoms concentration * 10 ^{12} (cm^{-3})
40	41	59	1.2
50	68	32	0.63
60	82	18	0.4
70	84	16	0.3
80	87	13	0.255
90	92	8	0.16
100	96	4	0.08
110	97.5	2.5	0.044
120	98.3	1.7	0.03

The measured values for the metastable atoms densities were found to lay from 1.2 × 10^{12} ± 10% cm^{-3} at 40 μs to 3 × 10^{10} ± 10% cm^{-3} at 120 μs.

As it can be revealed in the Fig. 3 (in the logarithmic scale representation) the $^3P_2$ Ne metastable density profile in the presented temporal range of the afterglow has an exponential decay with the decaying time resulting from the fitting curve of about τ_m = 22.6 μs. We may assumed that the main loss process in the investigated temporal range is the diffusion because the diffusion coefficient obtained considered the decaying time as diffusion time and diffusion length λ = 1 mm (lowest diffusion mode) is in very good agreement with the value reported by .V. Phelps [17]

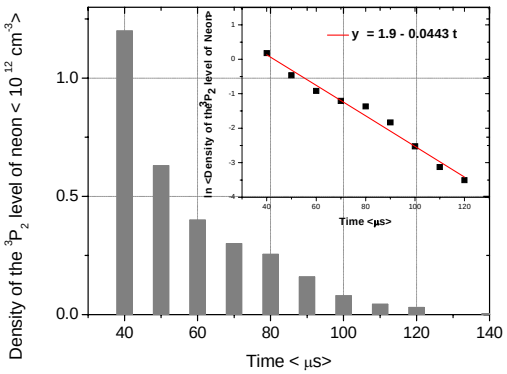


Fig. 3. $^3P_2$ Neon metastable level density profile according to the absorption measurements on 614.3 nm transition and $^3P_2$ level density profile in logarithmic scale.

4. Conclusions

The experimental spectroscopic technique presented in this paper is shown as a convenient tool for optical absorption spectroscopy with temporal resolution. The pulsed high current density spectral lamp emits intense spectral lines ensuring an appropriate signal/noise ratio for the absorption measurements. The analysis of the shock waves frequencies produced during the breakdown of a high voltage pulsed discharge allows evaluating and controlling the gas temperature and consequently the Doppler line profile in accordance to the discharge parameters.

By comparison with the more sophisticated laser absorption or laser induced fluorescence techniques, the technique of optical absorption spectroscopy with temporal resolution presented in this paper very useful for monitoring the processes occurring in the discharges working in pulsed regime is simple, inexpensive and easy to be implemented on different plasma reactors.

The high concentration, >10^{12} cm^{-3} of neon atoms in the metastable state (2^3P_2) at the 40 μs after the end of the high voltage pulse, indicates the fact that the temporal afterglow plasma of the hollow cathode pulsed discharge can be used as a supplementary energy source for excitation and ionization of sputtered atoms of samples in GDOES and GDMS analytical systems.

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