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Formation of low pressure striations in a krypton lamp, experimental characterization of the discharge: spectroscopic and electrical analysis

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Abstract:

The existence of striations in DBD - wave manifestation and oscillations of ionization - has long been an interesting topic, hampered in part by lack of empirical evidence. Striations are due to an alternation between regions of predominance of electron creation and regions of predominance of electron loss. This study gives a recent contribution to the existence of striations in a low pressure krypton plasma. Experimental analysis with an AC alternative current allowed us to distinguish two kinds of striations, mobile and immobile striations. In our case, the striations are formed one by one from the cathode towards the anode in a weak field channel. The formation of striations depends on the flow of modulated electrons. The photographs of emissions in this work were obtained by an intensified charge-coupled device (ICCD) camera with nanosecond exposure time. The manifestation of the striations was clearer at the naked eye and were more accurate with the camera. Spectroscopic and

electrical measurements are also considered in this estimation in order to study the behavior of the discharge, the characteristics of the energy deposit and its luminous efficiency. Then the striations in the Krypton lamp draw our attention and allow us to better understand the inhomogeneous aspect of the discharge.

Keywords: striations, pulsed discharge, inhomogeneous plasma, dielectric barrier discharge (DBD).

1. Introduction:

The first aim of this study was to improve the luminous efficiency of dielectric barrier discharge lamps. This upgrade was accomplished by understanding the mechanism of the discharges based on various noble gases (Ar [2], Kr). This contribution focused on the processes of conversion the electrical energy into light energy via the optimization of the physical parameters of the plasma (gas type, gas pressure, discharge gap, voltage amplitude and frequency, etc.) and the conditions of electrical excitation (excitation frequency, duty cycle and pulse duration).

Excimer lamps are good candidates to replace previous mercury-based lamps [3]. The UV produced by excimer lamps are also very important, recent study from Bernner's team report that far-UVC light kills pathogens in the air before we can breathe them in. Far-UVC wiped out coronaviruses responsible for coughs and colds, and it's assumed to be equally effective at liquidating the virus that causes COVID-19 [4].

In this paper, we focus specifically on the study in the case of a Krypton lamp to improve the performance of an excimer lamp (Kr_2^* at 148 nm [5]). The appearance of striations in our discharge led us to examine them experimentally using an iCCD camera. Plasma striation, as

an example of self-organization at the kinetic level, remains a considerable challenge for understanding the physics of gas discharges [6]. The physical explanation for this appearance remains inadequately comprehended, regardless of numerical investigations [7]. There are qualitative descriptions in simulations of moving striations, quantitative descriptions are expected to be obtained in the near future [8].

2. Experimental setup:

This part presents the experimental set-up and the diagnostic tools set up to characterize a plasma lamp. The experimental set-up (**Figure 1**) includes the equipment used to generate the plasma (plasma source, gas, power supply) and the diagnostic tools set up to study the lamp. The diagnostic tools are divided into two categories: electrical diagnostic tools (voltage probe Tektronix P6015A, current probe Pearson electronics 6585, oscilloscope Tektronix-TDS7104 digital phosphor) and optical diagnostic tools (iCCD camera, optical spectrometer ocean optics HR 2000+, optical fiber ocean optics SR 600 with diameter of 600 μm , the optical fiber was connected to a vacuum passage).

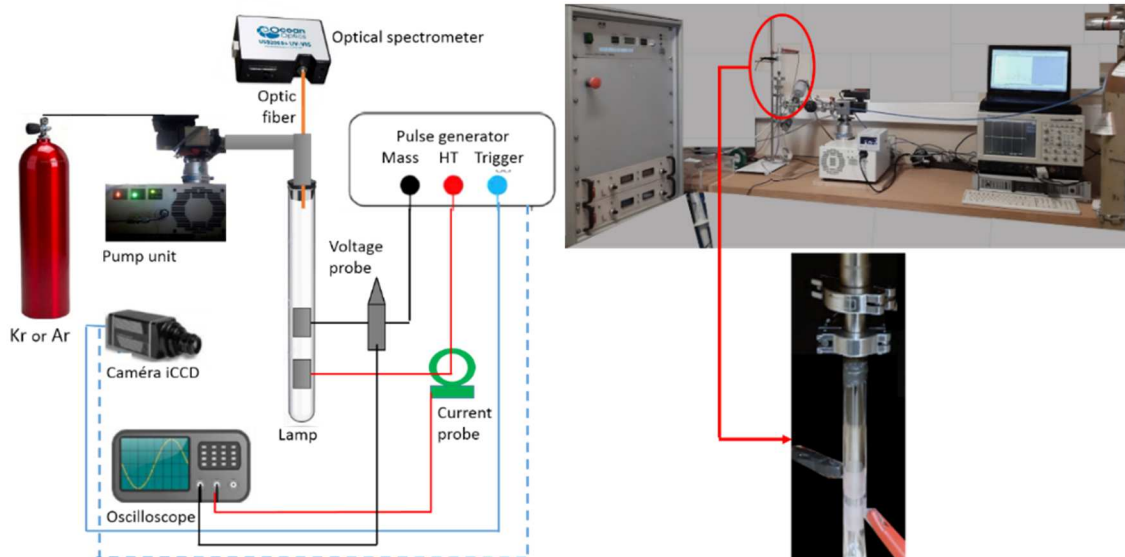


Fig.1: experimental device.

2.1 Plasma source :

The plasma source used in this work is a borosilicate glass lamp having a cylindrical geometry. The choice of glass is based on its isolator character being the only dielectric in the discharge, its hardness as well as its inert aspect. During the experiments, two lamps were used α and β . the α lamp was discussed in [2], the dimensions of β source are: 200 mm long, external diameter was 20 mm, glass thickness was 2 mm. The glass tube was fitted on a stainless steel flange fitting. Sealing is ensured by a polymer O-ring (viton seal) and Araldite glue.

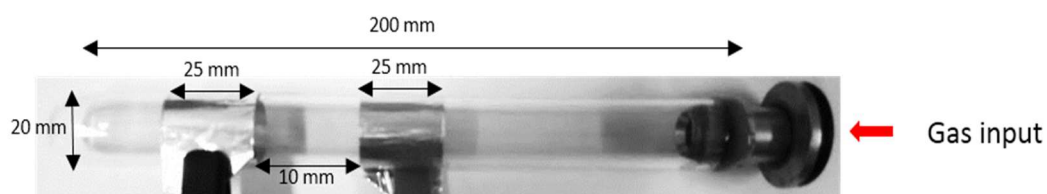


Fig.2: picture of the plasma source, β lamp.

The electrodes are made of aluminium (3M, tape 425, thickness 120 μm). The high-voltage was placed at the top of the source, while the ground electrode was at the bottom. The both, ground and high-voltage electrodes are positioned outside the lamp. The main interest of this discharge is that they used electrodes outside the plasma limiting their deterioration. The deterioration of the electrodes would induce pollution of the plasma made by species coming from the electrodes, this would cause a loss of luminous efficiency [9] [10].

2.2 Electrical diagnostics:

The power supply (A2e Technologies) allows the application of a sufficiently high potential difference between the two electrodes to create a plasma from the gas present in the lamp. The power supply is pulse type of high voltage giving a symmetrical square profile capable of providing a maximum voltage of 3 kV and a frequency from 10 kHz up to 100 kHz.

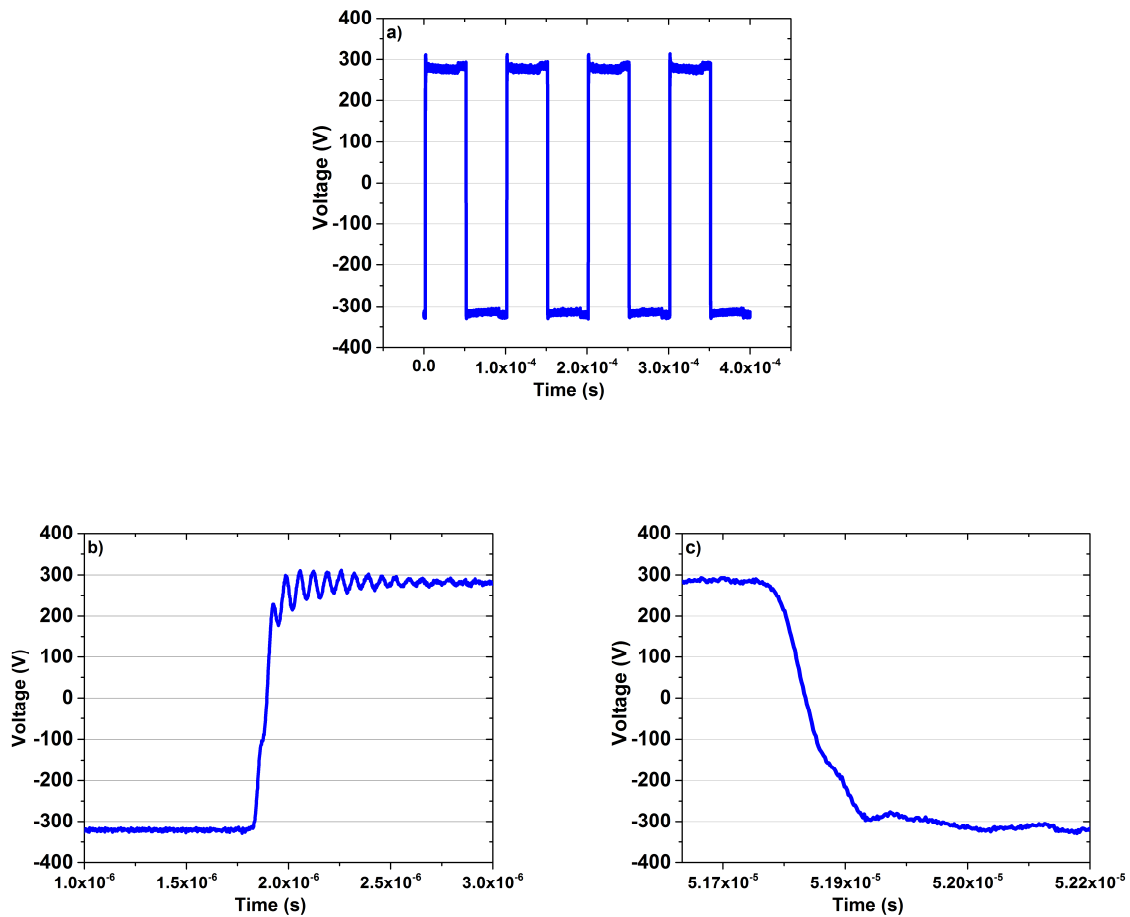


Fig.3: voltage signal measured at the discharge terminals for 280V - 20 KHz

a) on quarter periods, b) zoom on the rising edge, c) zoom on the falling edge of the pulse.

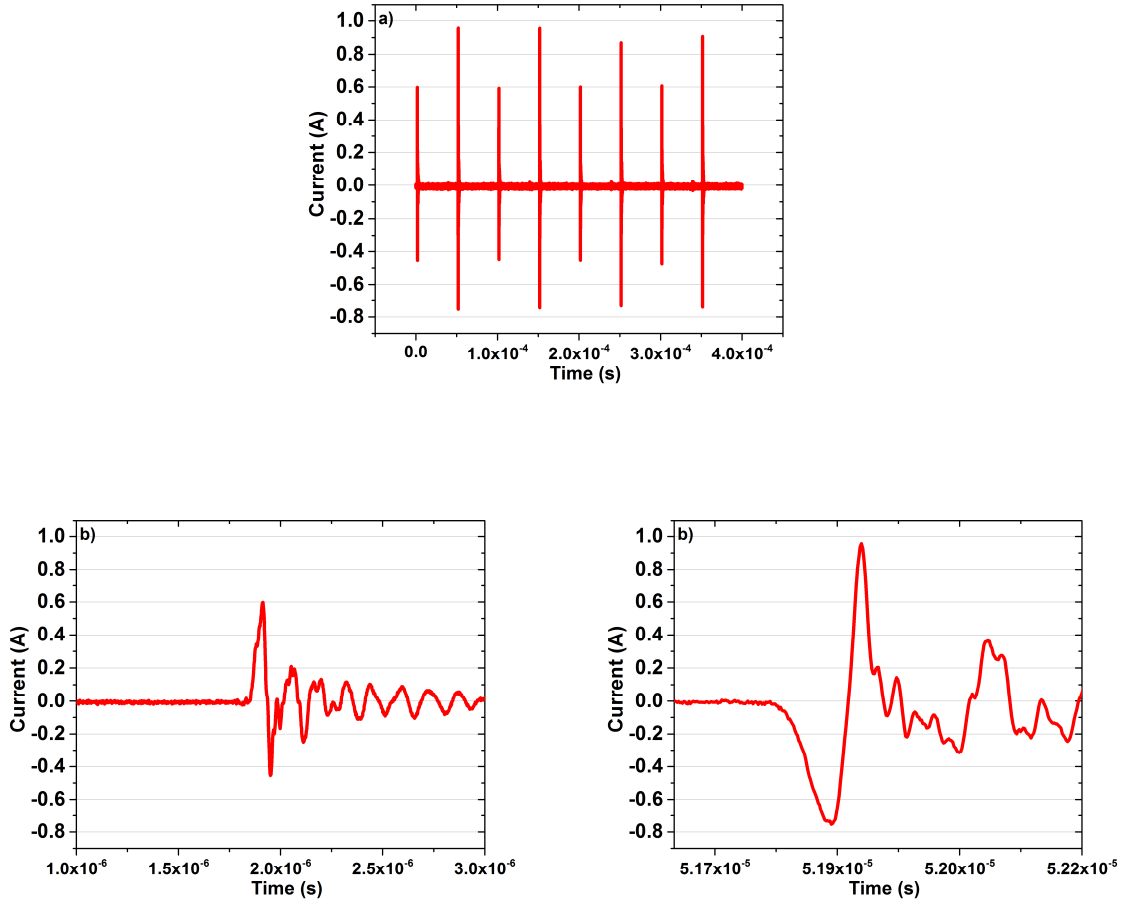


Fig.4: a) measurement of the discharge current over four periods, b) Zoom on the rising edge of the pulse, c) Zoom on the falling edge of the pulse, when the plasma is turned on.

2.3 Optical diagnostics:

The spectra were obtained using a spectrometer (Ocean Optics, HR2000+) operating in the wavelength range of 200 to 1100 nm and offering a best resolution of 0.35 nm. The spectrometer is coupled to an optical fiber (Ocean Optics, SR600) of diameter 600 μm . The optical fiber is connected with a vacuum passage to channel the plasma light. The optical spectra presented in **figure 5** are integrated spectra over times ranging from 100 to 600 ms, the pulse frequency was 20 kHz, the pressure was 60 mbar, applied voltage amplitude was

500 V with a gap of 1 cm on all spectra. The spectra in **figure 5** clearly showed the emission of the states of Argon and Krypton respectively. The presence of nitrogen, OH and oxygen on the Argon spectrum was partially the consequence of an electronic energy transfer between the metastable Ar (3P2) and the nitrogen molecule [2] [11].

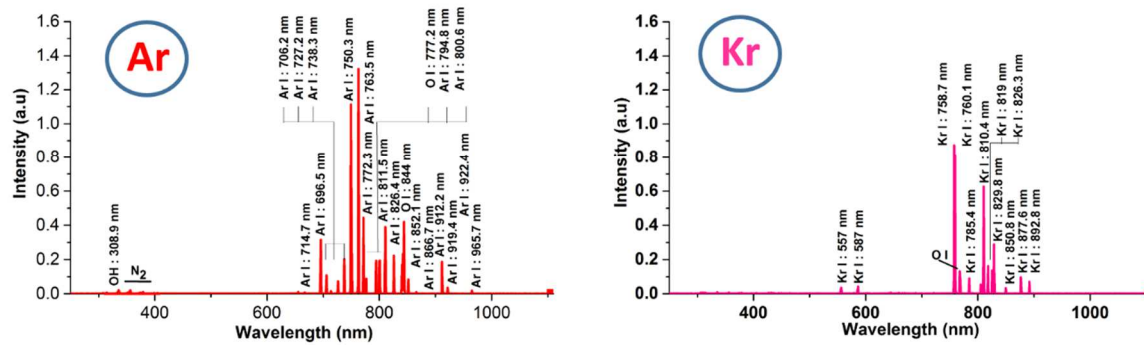


Fig.5: plasma spectra of Argon and Krypton respectively from the β lamp.

The iCCD (Intensified Charge Coupled Device) camera used in this study (Princeton Instruments PI-MAX) has a resolution of 1024*1024 pixels. The (Red-Blue) RB Intensifier provides a scope of wavelength quantifiable by the camera between 200 and 900 nm. A visible lens (Nikkor 80-400 mm, Nikon) is connected to the camera. The iCCD camera offers the possibility of accumulating plasma emissions over different periods of time. The frequency used for this manipulation was 20 kHz, the triggering of the camera is synchronized to the frequency of the discharge via an electrical signal (trigger) provided by the power supply as shown on the **figure 6**.

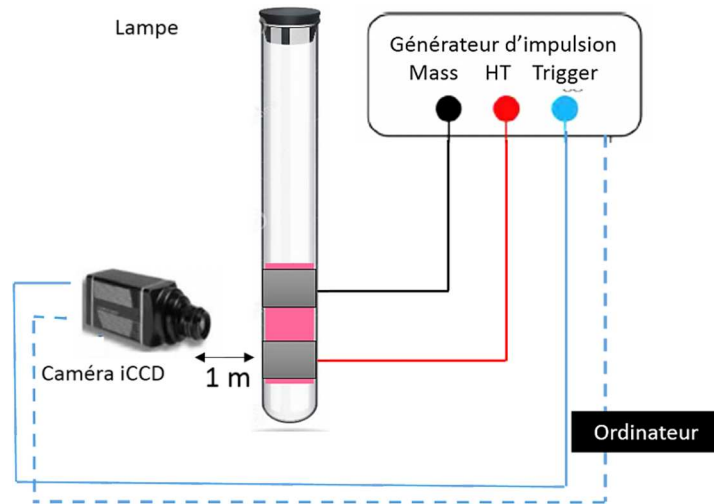


Fig.6: representation of the experimental device with the iCCD camera.

Figure 7 shows a sequence of photographs illustrating the temporal and the spatial evolution of the plasma during the positive transition of the voltage signal. When taking pictures the voltage and the pressure are kept constant, the plasma is formed and then the striations appear at different points of the gap, oriented up and down the center of the discharge space until their extinction. The striations are at the origin of the phenomenon of the organization of the discharge, the observation of these phenomena in DBDs using an iCCD camera are qualitative.

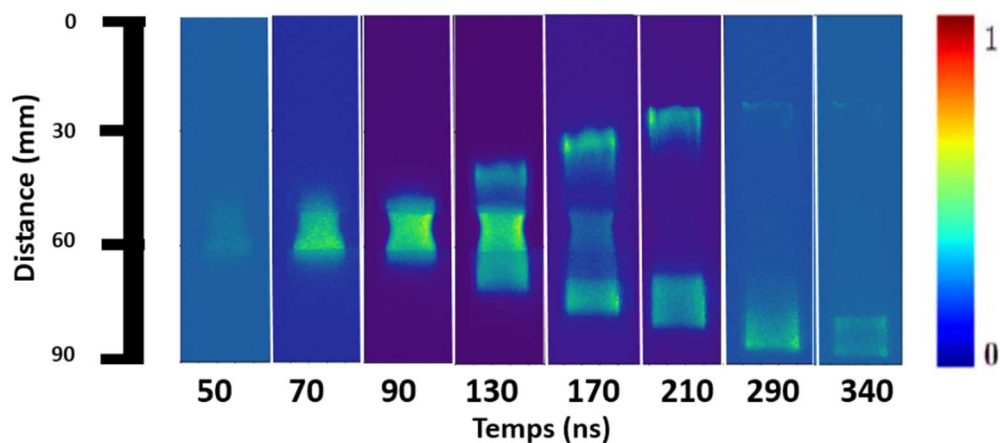


Fig.7: Krypton plasma dynamics in the lamp between the gap space during the positive transition of the voltage signal (30 mbar, 1 kV, 20 kHz).

Striations also affect the characteristics of the nature of a discharge (homogenous, inhomogenous plasma), this phenomena aren't desirable for most applications such as gas lasers and light sources [12]. In the design of fluorescent lamps, efforts are made to create conditions in which striations don't occur or can't be observed. In the case of this study, striations were observed with the naked eye. The use of the iCCD camera confirmed their existence, the perception of a waves kind moving and standing in the plasma and which disappears spontaneously. This is related to the concentration of certain reactive species in the plasma: the process of N_2 quenching of metastables Ar ($1s5$) in Argon plasma confirms that the metastables species are certainly one of the important sources in the formation of self organized striations plasma (SSPs) [13], this means that the Krypton metastables in our work play an important role in the formation of striations.

3. Calculation method using Argon gas:

Based on the electrical measurements of voltage and current, the average power is calculated with the python program using Simpson's method. Each of the electrical (current and voltage) and optical (spectra acquisition) measurements was performed three times. These three measurements are then averaged and the uncertainties are alculated as their standard deviations. The multi-value analysis was performed using the method << ANOVA one-way >> determined with OriginLab. In this case, the statistical significance p is set to 0.05.

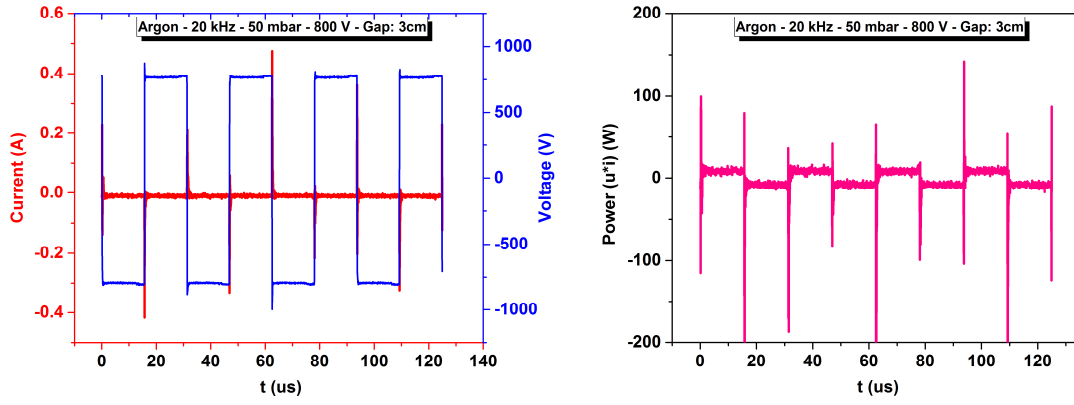


Fig.8: measurements of current, voltage and electrical power as a function of time.

The average power P_m is calculated with the following formula:

$$P_m = \frac{1}{T} \int_0^T u(t) \cdot i(t) dt \quad (\text{Eq.1})$$

P_m : average power in Watts, i : current applied in Ampere, u : applied voltage in Volt, T : period of discharge in seconds.

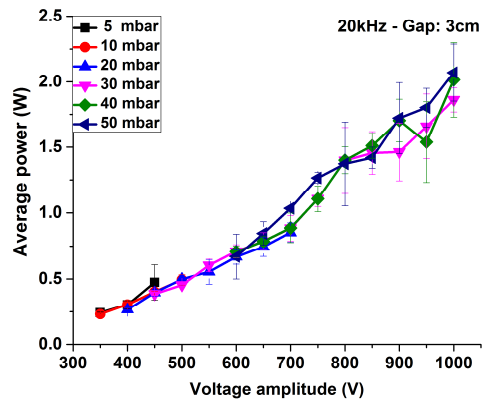


Fig.9: average power as a function of the voltage amplitude of Argon discharge.

Based on monitoring the maximum emission line of the gas we evaluate luminous efficiencies.

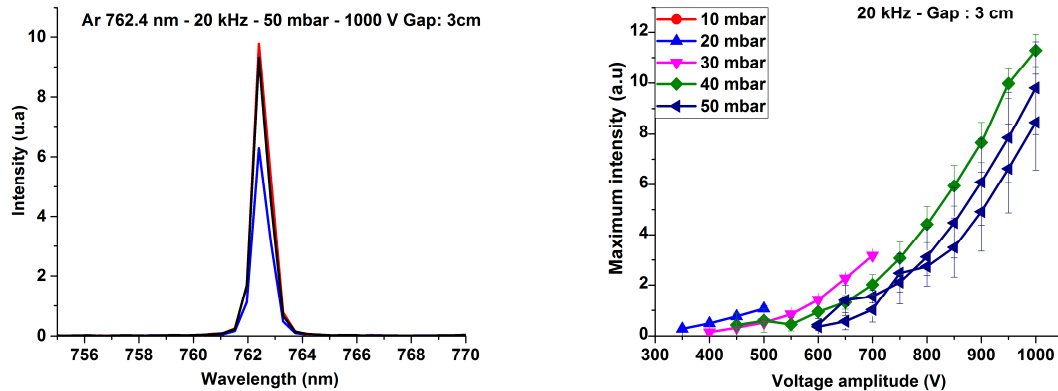


Fig.10: intensity of Ar 762.4 nm as a function of wavelength on the left, maximum intensity of the maximum Argon line at 762.4 nm as a function of the voltage amplitude on the right.

Luminous efficiency LE is defined as the ratio of the light output energy to the input energy [14]. The luminous efficacy is calculated from the maximum intensity of the gas under study. The maximum intensity is therefore divided by the integration time and the average power according to the following equation :

$$LE = \frac{I_{max}}{P_m \cdot t} \quad (\text{Eq.2})$$

LE: luminous efficiency, I_{max} : maximum intensity of the maximum line, t : integration time of the spectrometer, P_m : average power.

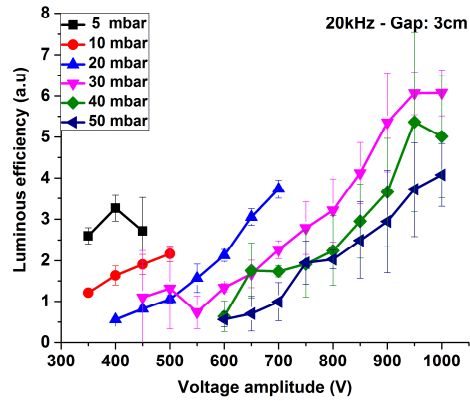
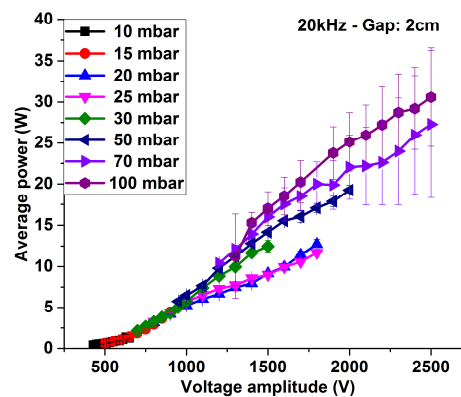
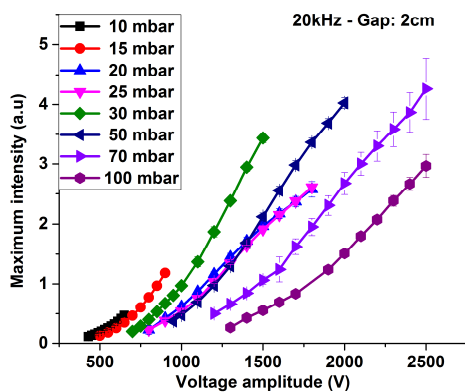


Fig.11: luminous efficiencies of the maximum Argon line at 762.4 nm as a function of the voltage amplitude of a typical discharge.

4. Results and discussion using Krypton gas:

The study was concentrated on best operating condition: gap conditions of 2 cm and at a frequency of 20 kHz. **Figure 12** shows the growth of maximum intensity, average power and luminous efficiency respectively as a function of the voltage amplitude. At **30 mbar - 1500 V** the luminous efficiency reaches its maximum and was greater than other measurements. At **p = 20 mbar – U = 1400 V** and **p = 25 mbar – U = 1700 V** there was an optimum of luminous efficiency. The curves of maximum intensity and average power increased with the voltage supplied, which is quite normal. But with regard to the graph of luminous efficiency, although it increased as a function of the voltage amplitude, the pressure curves did not follow the same order of growth as the other graphs, for example at **p = 15 mbar - U = 900 V** the luminous efficiency is better than at **p = 100 mbar - U = 2500 V**. There are a process that keeps the discharge from accomplishing a better efficiency despite the good energy deposit. In order to explain the process we used the iCCD camera, taking a measuring point **p = 30 mbar - U = 1000 V**, the point from which the luminous efficiency starts to grow to its

maximum. Visible and infrared camera analysis confirms us the existence of striations. Knowing that striations, stationary ionization waves were firstly describe in 1843 by Abria [15]. This phenomenon was probably already known to Faraday. Moving ionization waves (moving striations) were described in 1874 by Wullner [15], followed in 1876 by Spottiswoode [16]-[17]. Striations was also found in the AC coplanar plasma discharge panel research [18]. The plasma DBD is governed by different ionization processes (stepwise ionization, quenching of metastable, deviation of Maxwell's electron energy distribution and the heating of the gas) the instability of these processes causes the inhomogeneity of the plasma by the appearance of certain phenomena. The phenomena of striations in Krypton plasma are at the origin of an unstable ionization process causing the propagation of the waves. These waves cause the self-organization of the discharge, striation formation [13].



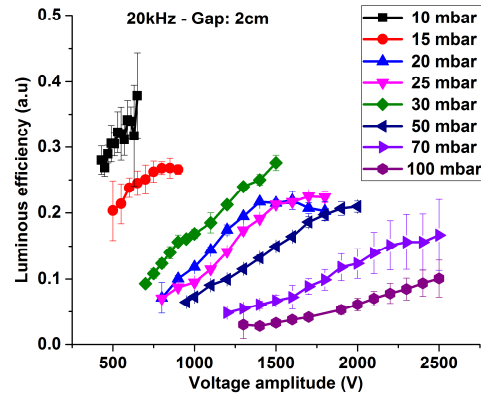


Fig.12: variation of maximum intensity, average power and luminous efficiency, respectively as a function of voltage amplitude at 20 kHz over a pressure range of [10 - 100] mbar, and at a gap of 2 cm (Krypton plasma).

The striations initially appear near the cathode and propagate towards the anode as reported by Sirghi in DC [19]. In **figure 13** the cathode is on the left, the anode is on the right. As reported by Kolobov [20] the negative light region forms near the cathode with a large peak in plasma density in the negative light region.

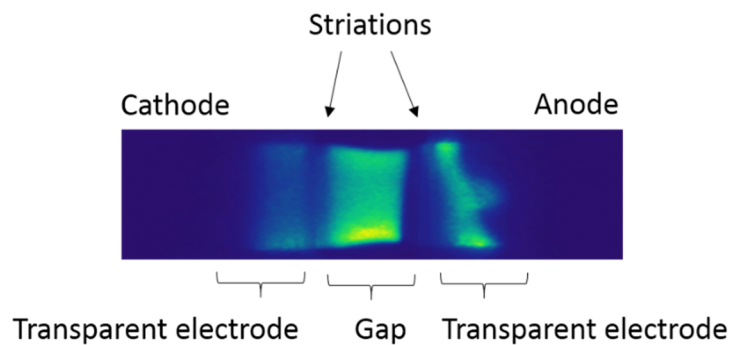


Fig.13: picture taken with the iCCD camera of the Krypton plasma between the gap when the striations appear (30 mbar, 1 kV, 20 kHz).

In **figure 14** the striations in a Krypton plasma move in the direction from the cathode to the anode (from bottom to top) between the gap and then dissipate, outside the gap area the striations don't exist. Striations split, merge and broad by changing the applied voltage [21]. Klyarfel'd [22] concluded from these experiments that there is no fundamental difference between stationary and moving striations, he considered them as a phenomenon with large amplitude. This kind of striations are periodic repetitions of a local perturbation in the plasma in the direction of the electron drift.

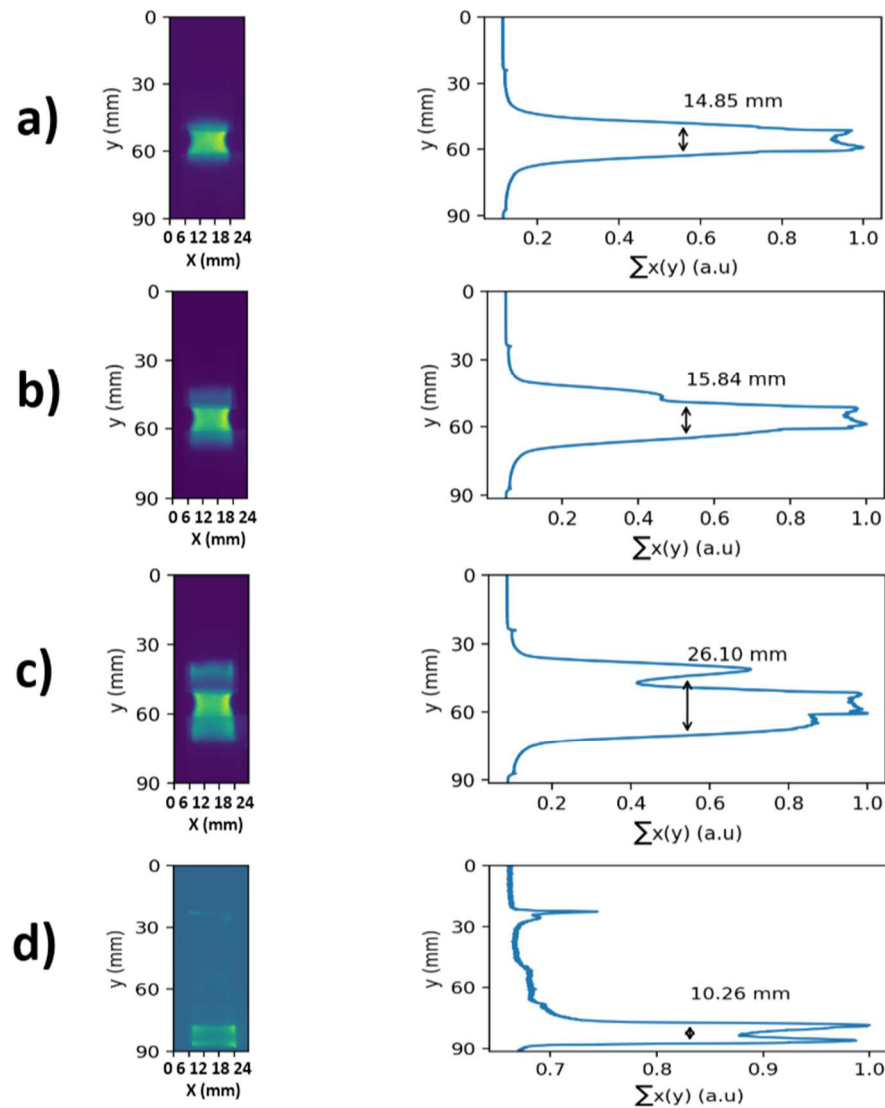


Fig.14: current profile obtained from the iCCD camera images between the inter-electrode gap during the positive transition of the voltage signal (30 mbar, 1 kV, 20 kHz).

4.1 Striations phenomena:

In DBDs, striations appear at variant pressures, from low pressure up to atmospheric pressure [23]. This study focuses on striations at low pressure, precisely at 30 mbar as show on **Figure 14**. The work was purely experimental. The work reported by Liu [1] concerning the evolution of the striations was a reference point. The hypothesis was that electrons moved from the cathode toward the anode because of the effect of electrostatic force, and obtained energy from the electric field for excitation and ionization process of Krypton atoms. Then, the positive space charges progressed towards the cathode, these are the occurrence of space charge at the place where the plasma homogeneity is locally disturbed by an excess of ions and electrons [15]. A strong electric field would be created near the cathode electrons will acquire enough energies at the edge of the positive column and striations are formed.

4.2 Types of striations:

There were two types of striations in the Krypton plasma, the stationary striations and the moving striations which moved from the cathode to the anode. The first study of moving striations was reported in 1920 by Aston and Kikuchi [24]. In 1962 Yoshimoto [25] specified that the positive moving striations in DC glow discharges are generated by the fluctuation of electron density and based on waves of the interaction between the space charge toward the cathode and electron density toward the anode [26]. Striations are also referred to as ionization waves because of they have a diffusion-ionization nature [27]. Both types of striations (mobile and immobile) are in the form of rings due to coaxial geometry of the lamp. The plasma takes the shape of the lamp and the shape of the electrodes which are responsible for supplying the energy.

In **Figure 14**, the plasma between the gap has a large peak of light intensity, this results comes from the strong processes that governs the positive column and are responsible for the formation of the plasma. In **(a)** and **(b)** photos the width of the light intensity peak increases as a function of time from **14.85 mm** to **15.84 mm** until two peaks are formed. In the **(c)** photo, there are two peaks of light intensity, the first peak was wider than the second peak and has a value of **26.10 mm**, the evolution of the light intensity of the second peak was not independent and was formed from the first peak of light intensity. The second peak is attached to the first peak, this could explain the mechanism of the striations despite of the formation of striations are not really understood [18]. The assumption is: when the peak of light intensity increases there is an important production of metastable, this leads to the appearance of striations. Knowing that the model proposed by Robertson indicated that with only a small number of metastable states then the discharge becomes stable [13]-[28] . In our case the discharge is not stable: the number of metastable states in Krypton plasma are higher than other states of other species and cause the apparition of striations.

The appearance of striations leads to conclude that the plasma is inhomogeneous, the light intensity of the plasma is not uniform, it is distributed differently on the gap space. In **(d)** photo, the width of the intensity pic starts to decrease until it reaches **10.26 mm** near the cathode, as the second peak is located at the end of the anode, the second peak is very narrow compared to its size in **(c)** photo, however the second peak seems to be detached from the first one until disappearance. The decrease in light intensity peak is probably caused by the loss of Krypton metastable by quenching reaction, the process of quenching of metastable Ar (1s5) confirms that the metastable is certainly one of the important sources in the formation of striations [13].

In general terms the striations start to form near the cathode when the electric field is strong enough, the striations accumulate charges and move towards the anode, this may be the

explanation for the appearance of the second peak of light intensity near the anode. Pekarek [15] explain in his work that a local increase of the density of the charged particles can't stay in the given place for a long time. There would be no wave phenomena, if the perturbation not lead to a change in the ionization rate per electron.

The striations confirmed that the AC discharge nature was not stable, despite the fact that striations are a brilliant example of self-organization at a typical kinetic level. Some progress has been achieved in understanding the nature of striations in classical DC discharges of rare gases [20], it has been confirmed that electron kinetics is responsible for the striation formation in most cases. This alternative can be adapting to AC discharge of this study, because of striation forming are similar in both pulsed DC discharges and DBDs [1]. Plasma is also estimated as spatially inhomogeneous medium and striations are considered as a particular form of self-organization observed in the discharge under various conditions of variation of the physical parameters [29].

5. Conclusion:

Excimer lamps remains the subject of much research. Ushio Inc company was the first of companies pioneering the technology of excimer lamp [30], knowing that excimer lamps opened entirely new possibilities in UV curing and in coating processes, in VUV surface cleaning, surface modification [30]. In general, UV are widely used in the applications of photophysics, photochemistry and photobiology. The first goal of this research was to optimize

an excimer lamp based on the Krypton gas for the production of UV and VUV via Kr_2^* , then the striations appeared in the β lamp and the focus was on explaining this phenomenon.

The optimization and the study of β lamp revealed that the discharge plasma is inhomogeneous. This phenomenon is explained by the disruption of the luminous efficiency results which were not consistent with the values of average power, but as far as this phenomenon is concerned, experimental description is not enough, a kinetic analysis must be added to this paper in future work. Advances must be made to better comprehension the role of kinetic reactions for the mechanisms of creation and loss of excited Krypton atoms, because it has been proven that moving striations appear when the excitation of Krypton gas is activated [31]. In addition, the use of fluidic models must be important to identify the basic mechanisms for the origin of these phenomena [32].

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