Experimentation and Modeling of Self-Organization in Cathode Boundary Layer Discharge in Noble Gas

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What is certain is not truth and what is uncertain is not ignorance.

Ilya Prigogine
Abstract

Self organized pattern formation (or self-organization) of microplasma in Cathode Boundary Layer Discharge is a phenomenon first seen in high purity Xenon (Xe) gas by research group led by Schoenbach at Old Dominion University [1]. Attempts by same research group to obtain similar results in other noble gases such as Krypton and Argon had failed. However, simulations performed by Pedro et. al. using COMSOL® suggested possibility of self-organization in Krypton as well as other noble gases at higher pressure [2]. Many competing models for the process of self organization were proposed [12].

At our laboratory, we have focused particularly in understanding the phenomenon of self-organization by gathering more data. Data were typically gathered by planar reactor structure in the pressure range between 50 Torr and 200 Torr, and such the results were used to evaluate the different competing models. In doing so, our experimental finding have verified some of the claims made in simulation by Pedro et. al. This includes the reporting of missing mode: the ring structure, which had not been previously observed, and the modes of structures leading upto ring structure [9]. Besides, self-organization was observed in Krypton and was found to be equivalent to the ones seen in Xenon [2] as suggested by the model. While Molybdenum was primarily used as the cathode material, cathode materials such as Aluminum, Hafnium, Tungsten, Silver, Steel, Nickel, Titanium, Zinc and Copper were also tested.

In addition, different reactor design, dielectric material, anode material and hole design were studied. In characterizing the plasma, the electrical properties of plasma were studied which included Current Voltage Curve (CVC), and Current Density Voltage Curve (CDVC). Additionally, optical emission spectrum of plasma were taken and studied carefully.
Acknowledgement

I started working on my research project since my sophomore year. Initially, it was the beautiful structures of the self-organization that got my interest. Later as I became more familiar with the concept of self-organization, I began to relish my research more deeply. My enthusiasm for the project only increased over time.

I would like to take this opportunity to thank my research advisor Dr. WeiDong Zhu for giving me a space in his lab. Without his support, motivation and guidance, I could never have conducted and completed the research for this thesis. His door was always open for discussions, and I cannot thank enough for all the laboratory skills that he has taught me over time. In addition, I would also like to thank Dr. Debing Zeng for his support and advice during my research. I am thankful to Dr. Pedro Almeida from University of Madeira, Portugal. His interesting discussions and insights into the nature of the problem, along with his simulation work has been a great source of help and information. Besides, I am also grateful for getting technical help from Dr. Gabriel Gomes whose short visit to our lab improved our experimental setting. I am also indebted to Bert Harrop from Princeton University, for giving me access to laser dicer which was helpful in preparing some samples in the experiment.

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Chapter 1

Plasma

1.1 Introduction

In physics, plasma is commonly referred as the fourth state of matter constituting the largest portion of the visible Universe. Unlike ordinary gas, plasma is in an ionized state where there are sufficient charged particles whose presence alters the properties and behavior of the gas [3]. It is formed by the dissociation of atoms or molecules into their positive and negative particles; thus while plasma is globally neutral, net local charges can be present. This property is referred as quasi-neutrality. Due to presence of positive and negative particles, plasma interacts with electromagnetic force, exhibiting properties such as conduction of electricity and magnetic confinement. However, not all components of plasma are ionized, and ionization degree is a physical term measuring the how many charged particles are there compared to total particles. Mathematically

\[ i = \frac{n_e}{n_t} \]  

(1.1)

Here, \( n_e \) is the average electron density, and \( n_t \) is the average atom or molecule density. This value is usually below or around 1%. Temperatures associated with plasma are usually higher than those associated with other condensed matter; and one of the ways of producing plasma is to heat a gas. This causes atoms to collide among themselves leading them to dissociate into positive and negative particles. Another way for plasma production is through the use of electric field. Any gas contains few freely occurring electrons due to photo-ionization or cosmic rays. Once strong enough electric
field is applied, the electrons upon collision with gas atoms or molecules lead to production of more electrons. This effect known as avalanche effect leads to development of sustaining channel for the flow of electrical charges, which in other words is plasma.

1.2 History of Plasma

Early form of plasma related experiment started as early as eighteenth century. These early experiments were conducted in discharge tubes, and led to discovery of cathode rays and phenomenon associated with it such as striations [3]. As shown in the figure, discharge tubes consisted of an evacuated glass cylinder with electrodes on the either side. Their progress depended upon the quality of vacuum that could be produced. Thus, as the better vacuum could be produced, better the experiments became. While presence of two different types of charges were known, it was only towards the end of nineteenth century, that J. J. Thompson correctly identified cathode rays actually consisted of electrons, and claimed them as “the fundamental quantity in terms of which all electrical processes can be expressed” [7].

![Different charge spaces in a traditional discharge tube.](image)

Also at the same time, altering dark spaces, negative column, positive columns and striations were observed and studied. Since the twentieth century, the range of experiments dealing with plasma widened rapidly. These included experiments to understand the effect of ionosphere in radio wave propagation, and use of gaseous electron tubes for rectification, switching and voltage regulation [4]. These studies played vital role in building of the first generation computers in the mid twentieth century. Following the Second World War, the race to produce fusion bomb led to more research in the plasma. Lately the focus has shifted to the production of a source of sustainable fusion energy through international collaboration as ITER (International Thermonuclear Experimental Reactor). These lines of
research have also led to better understanding about the working mechanisms of stars, and early stages of the Universe and its evolution.

1.2.1 The Term "Plasma"

The term plasma was already in use to describe blood plasma in biology when Langmuir, an American scientist, reused it in 1922, to describe the soup of electrons, ions and neutral atoms likening it with the fluid of the blood once the corpuscles were removed. However, this created confusion with the original usage of the term, and has sometimes been seen as a misnomer since there is no fluid medium involved in plasma like blood [4].

1.3 Types of Plasma

As the field broadened, plasma has been subsumed under two different categories based on temperature, thermal plasma and non-thermal plasma.

1.3.1 Thermal Plasma

As the name suggests, thermal plasma are typically very hot. Their state is characterized by Local Thermodynamic Equilibrium (LTE) in which to say that state is only the function of temperature [5]. The temperature of the plasma is usually higher than 5000 K, making it harder to contain in ordinary vessel. In addition, thermal plasma requires a large amount of energy to sustain. Ordinarily the thermal plasma would refer to fusion work. However, microplasma such as corona discharges also fall under this category.

1.3.2 Non-Thermal Plasma

On the other hand, no such equilibrium state is reached in non-thermal plasma, and energy are selectively fed to the electrons. Hence, electrons and gas are at two different temperature; the temperature of electrons is higher while the gas temperature do not rise much above the room temperature. This allows
cold plasma to be conveniently operated in normal condition, and has opened avenues in bio-medical applications.

Cathode Boundary Layer Discharge (CBLD) is an example of non-thermal plasma. It is subsumed under microplasma because the plasma discharges have dimensions in the range of a millimeter. The discharge is spatially confined between the electrodes. Initial experiments conducted by Schoenbach et. al. consisted of hollow cathode and hollow anode arc were referred to as Microhollow Cathode Discharges (MHCD). When a planar cathode was used instead of a hollow cathode, self-organization was observed for the first time. The discharge is limited to cathode fall, negative glow and anode fall while lacking positive column[6] [13] as seen in Fig 1.2.

Figure 1.2: Discharge in a typical Closed Boundary Layer Reactor.
Chapter 2

Experiment

2.1 Setup

The experimental setup consisted of a chamber where a plasma reactor was placed. The reactor consists of a planar cathode and open anode and annular dielectric, as shown in the fig (2.1). Both anode and dielectric have central holes of same diameter. For most part of our experiments we used pre-drilled silica with a diameter of 750 µm for dielectric, and the cathode and anode were primarily made of Molybdenum ($^{42}$Mo$_{96}$) having thickness 250 µm and are 99.95% pure obtained from Alfa Aesar®.

![Traditional Plasma reactor design.](image)

During the preparation of sample, both anode and cathode were punched out from metal sheet, filed and sanded with sandpaper of different grits of 150 and 240 so as to remove any sharp edges. Following sanding, the samples were sonicated in 30-50% concentrated acetone (by volume) for 3-5 minutes. Anode, dielectric and cathode were then assembled together using Torr Seal® and heated in oven at 120 °C for about ten-fifteen minutes. In course of preparation of plasma reactors, the cathode is especially handled with care, as self-organization was particularly sensitive to the cathode surface condition. The reactor was placed with a grounded aluminum holder, and was driven by a DC power source. The chamber itself is pumped using a combination of diaphragm and turbo-pump to a base
pressure of about 0.50 mTorr. Once pumped to the desired base pressure, the chamber was filled with
noble gas to a desired pressure. The DC power supply allowed to set the maximum current, which for
the most experiments was kept at 1 mA. Once at this stage, the voltage was increased until the device
ignited.

2.2 Schematics

Figure 2.2: A schematic diagram of the experimental setup.

The cathode of the plasma reactor in the reactor was connected to the negative polarity DC power
source (Glassman®), while the anode was grounded. Two digital multimeters were used to monitor
current and voltage respectively. The circuit consisted of 100 KΩ ballast resistance, and additional
potentiometer in series. High resistance potentiometer used for the purpose of experiment is one of the
distinguishing features from the past experiments, and were primarily aimed for better current steps
for the study of current voltage curve. However, they have also been identified as the cause of the new
modes of self organization observed in the experiments which can be found in Section 3.1.1.
2.3 Data Acquisition

In the initial phase of our experiments, we manually recorded the data (Current, Voltage, Pressure and picture). However, during the study of hysteresis and transition, it became important to have acquired the data faster and eliminate potential human error. Thus, optical character recognition method was employed for which opencv library in python was used. The algorithm looks for the contour of every digit, and employs seven segment character recognition. The core of the algorithm consisted of comparing average pixel of the greyscale image to the local average pixel for the positive identification. Since the domain space is small, the program is capable of taking 3-5 readings per second with an accuracy about 95%. With further error checks the accuracy can be improved. However, these accuracy were highly dependent on the lighting condition, and care had to be taken to avoid glare of the screen. There were also cases of digit misidentification, which were addressed by further error checks. The pictures of plasma were simultaneously taken using Panasonic GP-KR222 camera, a charged coupled device (CCD). These pictures were fed to the computer using Hauppauge® analog to digital converter and were later corresponded to data (current and voltage) by using time stamps. Since, it is not always possible to exactly match a picture with a data, or vice versa, method for manual recording of data was also built. A program was written in python to facilitate the research and allow smooth interface integrating different modules for the program.

2.4 Leakage

There is always a leakage into the chamber, which is known to inhibit the process of self-organization. Hence, it generally easy to obtain self-organization immediately after gas refill. Experiments in small pressure are more prone to effects of leakage as expected. Our chamber is roughly 400 cm$^3$ in volume. And leakage rate corresponding to 0.05 mTorr/min did not have a detrimental effect on the formation of self-organization. This roughly corresponds to $2 \times 10^{-7}$ml/s. Leakage higher by an order of magnitude makes it harder to obtain self-organization. At leakage higher by two orders of magnitude.
absolutely no self-organization was observed. In addition to leakage, there is also outgassing in the vacuum. However, pumping the chamber over a day is expected to decrease outgassing considerably, making it negligent in comparison to leakage itself. In order to get better base pressure, pumping while heating the chamber is also a good choice.
Chapter 3

Results and Analysis

In understanding the phenomenon of self-organization, roles of different parameters such as pressure, cathode material, geometry, dielectric material, dielectric spacing and reactor design were studied. Among different gases experimented with, Xenon was more rigorously studied, as past and our experiments have shown a more consistent observation of self-organization in Xenon. In addition to Xenon, Krypton and Argon were studied. Self-organization has been observed in Krypton [2], however has not been seen in Argon so far, although more rigorous experimentation would be required to conclude absence of self-organization. In addition, the simulations work, which are increasingly proven to be successful, posits that there ought to be self-organization in Argon. And the only reason that is thought as being responsible for absence of self-organization is the absence of rigorous experiments attempts.

3.1 Xenon

Xenon is the heaviest of the stable noble gas in the periodic table. It has been the gas of choice for the self-organization experiments, as past experiments have shown Xenon readily produce self-organization. In fact, it was the only gas in which self-organization was observed in CBLD before our experiment. For experiment, lecture bottles of research grade Xenon (purity 99.999%) was acquired from GTS Welco®.
3.1.1 Pressure

Different pressure translates to different particles concentration. For a typical CBLD system, different pressure gives rise to different self-organization patterns. No self-organization was observed below 40 Torr in traditional reactor. For pressure higher than 170 Torr, large number of spots were formed arranged in concentric circles while the size if the spots were observed to be smaller. For instance, for pressure in range of 60 Torr to 120 Torr, the spots size averaged 105 $\mu$m. For pressure around 350 Torr, the spots size measured were around 85 $\mu$m. Within the pressure range of 60 Torr to 120 Torr, the number of observed spots were directly proportional to the pressure i.e. higher pressure forming a higher number of spots. Also, it was observable that the central spot is larger than the spots on the ring. Clustered emission was observed at pressure as high as 760 Torr however, only in sub-atmospheric pressure range were the spots properly defined. In our study, The pressure that were primarily studied were between 50 Torr and 150 Torr. Higher pressure were particularly avoided because spots were not very distinct. In addition, higher pressure required in higher breakdown and higher operating voltage, which in turn damaged the plasma reactors quickly, and were therefore avoided.

During the experiment, when the breakdown voltage is reached a uniform discharge covering almost the entire cathode was seen. The voltage was then reduced, and with it the discharge reduced in size and moved away from the edge as seen in fig 3.1 and fig 3.2. Following this shrinkage of discharge, filamentation occurred developing homogeneous plasma into clustered emission points which were nearly identical both in shape and size. Initially, the filaments were not very distinct and are referred to as diffused modes. Further decreasing voltage led to formation of distinct spots, which appeared circular from the top view. Spots greater than 7 spots tended to form concentric layer of spots, and were seen to arrange themselves in regular polygon patterns. Further decrease in voltage decreased the number of spots usually one spot at a time. However, multiple spots can simultaneously disappear as seen in fig 3.2. It was also observed that in subsequent runs in experiment for a given current and voltage corresponding to a particular self organized state formed spots at the same locations, something similar to spatial memory effect seen in Dielectric Barrier Discharges (DBDs). However unlike DBDs
where memory effect occurs due to residual charges, here the spots location are pertinent to the hole condition and the cathode surface condition which is unique to each and every sample.

In both experimental run as depicted in fig 3.1 and fig 3.2, the last stable structure was a ring structure. We had consistently observed ring structure at different pressure. However, it was not seen in previous experiments [9]. The primary reason for the occurrence of ring structure has been reasoned as the addition of two high resistance potentiometers in series. The potentiometer not only allowed finer tuning of current, but also changed the external circuitry which is suspected to play role in states reached. Not only the ring structure was observed, but also the elongation of circular spots in the tangential direction which ultimately formed a ring. The ring structure had also been seen to form from two,
three, four or five spots situation, higher pressure leading to formation of ring with higher number of spots. Fig 3.3 shows the evolution of spots from five spots. It is interesting to note that current does not change noticeably during elongation of spots through all the process. When left to themselves, the elongated spots were observed to return back to circular spot situation. [9]. However, in other experiments, we have also observed such structures to be stable. While in both of the experimental runs shown in Fig 3.1 and Fig , ring structure was the ultimate stable structure sometimes it would further evolve into a single spot situation. The possibility of ring structure as a spot revolving around had been considered. Similar pattern formed in Dielectric Barrier discharge has been reported, and time resolved picture helped to conclusively prove the loop structure arising as a result of rotation of traveling filaments [10]. Similarly, using a high speed camera would allow us to conclusively see if ring structure arises from the rotation of the spots, which is in one of the future target in our research. So far ring structure has been observed in reactors made of Molybdenum and Tungsten as cathode material.

Figure 3.2: Self-organization observed in Molybdenum cathode at 100 Torr.
Fig 3.3 shows current-voltage curve at five different pressure. There are essentially three portion of the IV curve. First region is characterized by linear interdependence of voltage and current. This is the region of homogeneous discharge, which is referred as the region of abnormal discharge. In this phase, the plasma shrinks in size and detaches from the boundary. It however is of little interest in the study of self-organization. The next branch is characterized by larger slope, which begins with filamentation and corresponds to formation of distinct spots. The transition from abnormal glow into the region of filamentation is characterized by negative differential resistance, which is prominent in higher pressure as seen in the current voltage graph in fig 3.4. The data were taken with the help of optical recognition method described in the data acquisition section.
Figure 3.4: IV Curve at different pressure of CBLD.
Figure 3.5: Current Density Voltage curve at 75 Torr Xenon along with corresponding Current Voltage Curve

Fig 3.5 juxtaposes Current Density Voltage Curve (CDVC) with Current Voltage Curve (CVC). While the latter is almost monotonic, the former has a distinct minimal turning point denoted by point (b). Study of the corresponding point in Current Voltage graphs shows that the critical point in the CDVC corresponds to the onset of filamentation. After filamentation, the plasma relatively stabilizes the current, while the value of voltage decreases. This causes rise in current density in the subsequent modes of self-organization. Current density value has been measured using a program which counts the number of pixels above a specified threshold value, and converts this value into relevant area by using a scale factor. While this threshold value does not have much of an effect for the plasma in abnormal region, spots situation and ring structure are very sensitive to this threshold value. Thus, the current density for the fluctuates strongly based on this value. For instance, the loop structure in the CDVC is
denoted by (d) would have much lower value if a higher threshold value is used, whereas in comparison spot situation (c) would vary slightly, and the abnormal discharge denoted by (a) would be minimally affected.

![Figure 3.6: Pixel Values calibrated against the electrical power.](image)

The camera used in the experiment is a Panasonic GP-KR222. When the aggregate pixel is plotted against the electrical power, a linear relationship is seen as expected, thus allowing us to conclude that the pixel value is representative of the radiant energy. The positive y-intercept is accounted by considering the contribution by noise. Using the aforementioned assumption, the value of spots along its diameter is taken for red, blue, green, and total values, and are plotted in Fig 3.7. The green value saturates towards the center of the spots, while both red and blue reach maximum towards the center. Roughly speaking, the decay of the pixel intensity is exponentially proportional to its distance from the center. Hence, the spots are not uniform islands of emission but has a core from which it diffuses.
3.1.2 Cathode Material

During the experiments, Molybdenum has been primary choice for both anode and cathode. Experiments were also conducted with Copper, Steel, Hafnium and Nickel. Among these materials, copper did not produce self-organization. Steel, Hafnium and Nickel were shown to produce self-organization, however unlike Molybdenum it was only after multiple trials was the self-organization observed. The work functions of the metals are presented below for comparison [14]. A quick comparison shows metals producing self-organization and those not producing does not have significantly different work function values. To explain the variation in experimental results, electron emission processes has been considered, the discussion about which can be found in the theory and simulation section.
<table>
<thead>
<tr>
<th>Element</th>
<th>Work Function (eV)</th>
<th>Self-Organization</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aluminum (Al)</td>
<td>4.28</td>
<td>Not Observed</td>
</tr>
<tr>
<td>Copper (Cu)</td>
<td>4.65</td>
<td>Not Observed</td>
</tr>
<tr>
<td>Hafnium (Hf)</td>
<td>3.9 ± 0.1</td>
<td>Observed</td>
</tr>
<tr>
<td>Molybdenum (Mo)</td>
<td>4.6 ± 0.15</td>
<td>Observed</td>
</tr>
<tr>
<td>Silver (Ag)</td>
<td>4.26</td>
<td>Observed</td>
</tr>
<tr>
<td>Titanium (Ti)</td>
<td>3.96 ± 0.04</td>
<td>Observed</td>
</tr>
<tr>
<td>Tungsten (W)</td>
<td>4.55</td>
<td>Observed</td>
</tr>
<tr>
<td>Zinc (Zn)</td>
<td>4.33</td>
<td>Not Observed</td>
</tr>
</tbody>
</table>

**Table 3.1 Work functions of different cathode materials**

In addition to cathode materials, anodes were changed, however, no difference had been observed during the use of different anode materials. Also the dielectric of twice the thickness (500 µm) were used, and the patterns observed in such reactors were not noticeably different.
Figure 3.8: Self-organization as observed in Hafnium at 100 Torr Pressure.
Figure 3.9: Self-Organization in Steel at 130 Torr Pressure.
3.1.3 Design

Figure 3.10: Two new designs of reactors: with additional top layer of dielectric and with additional layer of annular cathode

The primary design used during the experiment was closed plane microcathode which has been described in section 2.1. In addition, two different designs were experimented which are shown in figure 3.9. The first design had an additional dielectric on the top. Reactors with top dielectric layer did not change nature of self-organization in Xenon. The other reactor design consisted an additional layer of cathode, which are referred to as closed plane microcathode, [8] but may better be referred as cathode well reactors. It produced completely new patterns of self-organization as shown in figure 3.11 and figure 3.12. Unlike the traditional discharges, these discharges were not uniform to begin with. They initially formed a ring of plasma. Even at higher voltage, the plasma did not cover the cathode surface, rather there was a gradient of plasma from edge towards the center. When the voltage was reduced, filamentation of plasma set in. This with further decrease of voltage led to formation of distinct spots. The spots, unlike those seen in traditional reactors, were beans shaped with convex region towards the edge. At 70 Torr, novel structures were seen as reported in fig 3.11.
Figure 3.11: Pattern formation in Cathode Well Reactor at 70 Torr.

Figure 3.12: Formation of novel self-organized states at 50 Torr Xenon in Cathode Well Reactor.
Figure 3.13: Current Current-Density Voltage curve at 60 Torr as observed in
The CVC exhibits similar properties as the traditional CBLD. The initial linear current voltage relation likens to the abnormal region where there is a linear dependence of voltage on current. However unlike traditional cathodes where plasma covers all of the cathode surface, increasing current leads to increasing size of plasma discharge until it completely covers the cathode surface. With the beginning of filamentation current relatively stabilizes while voltage continues to decrease. Although there is a lack of a distinct turning point as in a traditional CBLD, the lowest value of current density reached is comparable to values obtained for a traditional cathode.

In addition to aforementioned structures, non-standard holes such as square holes and triangular holes were studied. Making such hole required having an access to laser system which could cut through metals with a precision. For our experimental purpose, we used Nd:YAG laser which have a wavelength of 1064 nm. During the sample preparation, alumina of thickness 250 µm was attached to anode which consisted of a single layer of Molybdenum using Torr Seal. The right shape and right size hole was cut through the material by laser. Once the hole was made, it was sonicated in 30-50% concentrated acetone solution (by volume), after which cathode was assembled using the standard procedure using Torr Seal.

In a square hole, taking into consideration the distortion which is caused by debris at the edges, the plasma arrange in are rotationally symmetrical fashion (fig 3.14 and fig 3.15). However, since the debris on the wall was not as clean as circular samples the patterns appeared distorted from the expected symmetry. We made interesting observations at 80 Torr when the spots were found to arrange in a rectangular grid fashion (fig 3.15). However, at lower pressure, while the initial filamentation process created patterns unique to the sample, the self-organized with distinct spots were not very different from patterns already observed in traditional structures. Similarly in triangular holes (fig 3.17 and 3.18), new patterns were seen, generally exhibiting the rotational symmetry. Both of the design followed the general pattern of self-organization: filamentation preceding self-organization itself. However, one of the things that highlights the difference between square and triangular reactors against traditional form of self-organization was the spot location. Unlike traditional cathode, the spots tend to form at the
edges first (fig 3.15 and fig. 3.18).

The current voltage curve and current density voltage curve was studied. Similar to the traditional cathode reactors, initial current voltage were linearly interdependent before the beginning of the filamentation, after which there is relative stabilization of current. Also like in traditional reactors, current density voltage curve we have to a reversal in current density after the initial filamentation. One of thing that has not been studied properly in this research is the effect of area on the plasma formation and its subsequent effect on the current density. The square sample had dimension of 750 µm, so did the triangular sample. Because the samples started with different hole size, thus a comparative study of current density is not done. Over all, the study has shown the shape of hole affects the plasma in the initial phase of filamentation as well as subsequent arrangement of the spots, although the influence on the former is more distinct in the abnormal mode of operation. The rotational symmetry was retained in all of the scenarios. In addition, samples with larger inter-electrode separation (500 µm and 750 µm) were used. However, no interesting results were seen for reporting.

Figure 3.14: Spots arrangement in a square hole at 50 Torr.
Figure 3.15: Spots arrangement in a square hole at 80 Torr.

Figure 3.16: Current Voltage Curve and corresponding Current Density Voltage Curve at 50 Torr.
Figure 3.17: Spots arrangement in a triangular hole (dimension 750 µm each) at 65 Torr.
Figure 3.18: Spots arrangement in a triangular hole (dimension 750 µm each) at 100 Torr.

Figure 3.19: Current Voltage Curve and corresponding Current Density Voltage Curve at 65 Torr.
3.1.4 Hysteresis

CBLDs are non-linear systems. Thus, it is important not only where a state occurs but how it is reached. Such non-linear behavior could be visually confirmed as different organization patterns would be seen when decreasing the voltage versus when increasing the voltage. This was further confirmed by the study of CVC where hysteresis was observed. Hysteresis is particularly prominent in the region where filamentation begins. It was experimentally observed that between 75 to 120 Torr, higher pressure produces more distinct hysteresis as can be seen in fig 3.13. Current voltage when decreasing voltage and when increasing voltage are plotted using different color for comparison. The probable cause for hysteresis are described in the theory and simulation section.

![Figure 3.20: Observation of Hysteresis at different pressure in Xenon.](image-url)
The spectrum of Xenon was recorded with the help of tabletop spectrometer from Newport (Model OSM-400-UV-NIR) which has a resolution of 1 nm. The range of the spectrum that can be observed with the spectrometer is from 200nm-1100nm. The strongest emissions occur in the infrared region. Spectral lines in visible region were less prominent, and in UV region were almost non-existent primarily because they were absorbed by the atmosphere. In following table the observed spectral lines are matched with the spectral lines obtained from the NIST database [16]. All of the spectral lines can be accounted by Xe I discharge alone.

![Figure 3.21: Spectrum of Xenon Discharge at 100 Torr Pressure.](image)
<table>
<thead>
<tr>
<th>Observed</th>
<th>Corresponding</th>
<th>Transition</th>
<th>Energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>468.5 nm</td>
<td>469.1 nm</td>
<td>$5p^5(2P_{3/2})6s - 5p^5(2P_{3/2})6p$</td>
<td>$8.32$ eV - $10.96$ eV</td>
</tr>
<tr>
<td>824.5 nm</td>
<td>823.16 nm</td>
<td>$5p^5(2P_{3/2})6s - 5p^5(2P_{3/2})6p$</td>
<td>$8.31$ eV - $9.82$ eV</td>
</tr>
<tr>
<td>829.5 nm</td>
<td>828.01 nm</td>
<td>$5p^5(2P_{3/2})6s - 5p^5(2P_{3/2})6p$</td>
<td>$8.43$ eV - $9.93$ eV</td>
</tr>
<tr>
<td>883.4 nm</td>
<td>881.9 nm</td>
<td>$5p^5(2P_{3/2})6s - 5p^5(2P_{3/2})6p$</td>
<td>$8.31$ eV - $9.72$ eV</td>
</tr>
<tr>
<td>896.9 nm</td>
<td>895.2 nm</td>
<td>$5p^5(2P_{3/2})6s - 5p^5(2P_{3/2})6p$</td>
<td>$8.43$ eV - $9.79$ eV</td>
</tr>
<tr>
<td>906.4 nm</td>
<td>904.5 nm</td>
<td>$5p^5(2P_{3/2})6s - 5p^5(2P_{3/2})6p$</td>
<td>$8.31$ eV - $9.82$ eV</td>
</tr>
<tr>
<td>918.4 nm</td>
<td>916.3 nm</td>
<td>$5p^5(2P_{3/2})6s - 5p^5(2P_{3/2})6p$</td>
<td>$8.43$ eV - $9.79$ eV</td>
</tr>
<tr>
<td>982.2 nm</td>
<td>979.7 nm</td>
<td>$5p^5(2P_{3/2})6s - 5p^5(2P_{3/2})6p$</td>
<td>$8.31$ eV - $9.58$ eV</td>
</tr>
<tr>
<td>994.7 nm</td>
<td>992.3 nm</td>
<td>$5p^5(2P_{3/2})6s - 5p^5(2P_{3/2})6p$</td>
<td>$8.43$ eV - $9.69$ eV</td>
</tr>
<tr>
<td>896.9 nm</td>
<td>895.2 nm</td>
<td>$5p^5(2P_{3/2})6s - 5p^5(2P_{3/2})6p$</td>
<td>$8.43$ eV - $9.79$ eV</td>
</tr>
</tbody>
</table>

Table 3.2 Identification of spectra lines of Xe Plasma at 100 Torr
3.2 Krypton

Takano et. al. considered Krypton as the second best candidate for observation of self-organization given its position in the periodic table, yet no self-organization was observed in their attempts. However, as expected self-organization has been observed in our study in Krypton[2].

3.2.1 Pressure

The observed patterns for self-organization varied with pressure as expected. As in Xenon higher pressure typically led to formation of higher number of spots. The lowest pressure at which self-organization was observed was 105 Torr, while clustered patterns had been seen at pressure as high as 1300 Torr. The optimum pressure range for the study of self-organization is between 105 - 300 Torr. Like in Xenon, the discharge current initially covers the cathode surface. Decreasing the plasma leads to filamentation of plasma, which ultimately leads to development of distinct plasma spots. Fig 3.15 and fig 3.16 show evolution of plasma with decreasing current at 150 Torr and 240 Torr. Some of the structures observed in Krypton were rather interesting, and were never seen in Xenon. However, these patterns were not repeatable, making it difficult for the detailed study.
Figure 3.22: Self-organization in Krypton CBLD at 150 Torr along with corresponding current and voltage.
Figure 3.23: Self-organization state in Krypton CBLD at 240 Torr along with corresponding current and voltage.
The CVC and CDVC shows similar traits as those shown by Xenon discharge. Initially the plasma operates in the abnormal region where current voltage are linearly dependent. At this stage the discharge covers almost all of the cathode surface. With the onset of filamentation, current relatively stabilizes when the voltage drops off steeply. This also corresponds to the turning point for the current density, after which it begins to increase. In comparison to Xenon negative differential conductivity appears more pronounced, which could be one of the reasons for less frequent observation of self-organization in Krypton. Also the critical current density observed at the turning point is comparable with the critical current density observed in Xenon.

3.2.2 Cathode Material

Different cathode material were used with Krypton discharge. However, no self-organization were observed in cathodes except for Molybdenum. The empirical probability of obtaining self-organization in Krypton is low compared to that in Xenon, and since the probability of observing self organization
in cathodes other than Molybdenum is low, the combined probability for obtaining self-organization in Krypton in non-Molybdenum cathode would empirically be very low. More extensive research and better theoretical modeling would be required to conclusively explain the absence of self-organization in these different combinations. Research along these lines would help to find the role of secondary emission coefficient in the formation of self-organization.

3.2.3 Design

Reactors with top dielectric layer were experimented with Krypton. Initial experiments showed such addition favored the formation of self-organization in samples with an additional top layer of dielectric as well as polished cathode surface. However, later experiments showed possibility of obtaining self-organization in reactors both without top dielectric surface, and unpolished surface. Top dielectric layer is thought to assist in self-organization by increasing the electric retention through repulsion. The newer reactor design with cathode well structure as well as square and triangular hole were also tried in Krypton. However, no self-organization was observed. It has been observed that that obtaining self-organization in Krypton is experimentally harder than obtaining in Xenon, and requires operation at higher pressure, which in turn is also not favorable for formation of self-organization in CBLD. However, in the light that in traditional reactor design Krypton and Xenon are found to be completely equivalent, it is assumed that plasma in Krypton should also be able to self-organize even in newer reactor designs.

3.2.4 Spectrum

The spectrum of Krypton was also recorded with the help of tabletop spectrometer from Newport Model OSM-400-UV-NIR. The strongest emissions were observed in the infrared region, while some spectral lines were observed in visible region however were weaker, and in UV region spectral lines were almost non-existent. All of the spectral lines were accounted for by Kr I species ([16].
Figure 3.25: Spectrum of Krypton Discharge at 160 Torr.
<table>
<thead>
<tr>
<th>Observed</th>
<th>Corresponding</th>
<th>Transition</th>
<th>Energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>433.5 nm</td>
<td>431.96 nm</td>
<td>$4s^24p^5(2P_{3/2}^o)5s - 4s^24p^5(2P_{3/2}^o)5p$</td>
<td>9.91 eV - 12.8 eV</td>
</tr>
<tr>
<td>558.6 nm</td>
<td>557.0 nm</td>
<td>$4s^24p^5(2P_{3/2}^o)5s - 4s^24p^5(2P_{3/2}^o)5p$</td>
<td>9.91 eV - 12.1 eV</td>
</tr>
<tr>
<td>588.9 nm</td>
<td>587.1 nm</td>
<td>$4s^24p^5(2P_{3/2}^o)5s - 4s^24p^5(2P_{3/2}^o)5p$</td>
<td>10.0 eV - 12.1 eV</td>
</tr>
<tr>
<td>761.8 nm</td>
<td>760.2 nm</td>
<td>$4s^24p^5(2P_{3/2}^o)5s - 4s^24p^5(2P_{3/2}^o)5p$</td>
<td>9.92 eV - 11.5 eV</td>
</tr>
<tr>
<td>770.6 nm</td>
<td>768.5 nm</td>
<td>$4s^24p^5(2P_{1/2}^o)5s - 4s^24p^5(2P_{1/2}^o)5p$</td>
<td>10.6 eV - 12.3 eV</td>
</tr>
<tr>
<td>770.6 nm</td>
<td>769.45 nm</td>
<td>$4s^24p^5(2P_{3/2}^o)5s - 4s^24p^5(2P_{3/2}^o)5p$</td>
<td>9.92 eV - 11.5 eV</td>
</tr>
<tr>
<td>812.8 nm</td>
<td>810.4 nm</td>
<td>$4s^24p^5(2P_{3/2}^o)5s - 4s^24p^5(2P_{3/2}^o)5p$</td>
<td>9.92 eV - 11.4 eV</td>
</tr>
<tr>
<td>820.5 nm</td>
<td>819.0 nm</td>
<td>$4s^24p^5(2P_{3/2}^o)5s - 4s^24p^5(2P_{3/2}^o)5p$</td>
<td>10.0 eV - 11.5 eV</td>
</tr>
<tr>
<td>827.7 nm</td>
<td>826.3 nm</td>
<td>$4s^24p^5(2P_{1/2}^o)5s - 4s^24p^5(2P_{1/2}^o)5p$</td>
<td>10.6 eV - 12.1 eV</td>
</tr>
<tr>
<td>879.3 nm</td>
<td>877.7 nm</td>
<td>$4s^24p^5(2P_{3/2}^o)5s - 4s^24p^5(2P_{3/2}^o)5p$</td>
<td>10.0 eV - 11.4 eV</td>
</tr>
<tr>
<td>894.5 nm</td>
<td>892.9 nm</td>
<td>$4s^24p^5(2P_{3/2}^o)5s - 4s^24p^5(2P_{3/2}^o)5p$</td>
<td>9.9 eV - 11.3 eV</td>
</tr>
</tbody>
</table>

Table 3.3 Identification of spectra lines of Kr Plasma at 160 Torr
3.3 Comparison between different gas

Figure 3.26: Equivalent stages of pattern formation as seen in Xenon and in Krypton. The blue discharge are from Xenon while the red are from Krypton.

It was shown that almost all different patterns of self organization produced in Xenon can also be reproduced in Krypton at pressure almost twice as high. This includes structures seen with the onset of filamentation, distinct spot situation with one, two, three to multiple spot situation. Multiple concentric circles spot arrangement as well as ring structures were also observed in both Krypton and Xenon. This result helped to establish that Krypton Discharge and Xenon discharge are equivalent to one another.

Experiments were also conducted with Argon, however it failed to produce any self-organization. CVC curve shows the plasma operating in abnormal region. The plasma decreased in size with when the current was decreased and finally extinguished. No filamentation and clustered emission was observed. The plasma operated only in the abnormal region, and fails to move jump from abnormal mode.
3.4 Surface Modification

The surface of cathode is continuously bombarded by ions. When the ions have energy that is greater than the lattice displacement energy, they can displace atoms from the crystal structure. On the other hand, collisions can also lead to emission of x-rays, optical photons and electrons. [15] A quick calculation shows that the ions have energy less than 1 eV, far less than required to modify the surface. Yet, as noted in the earlier in presenting the results, the breakdown voltage of a reactor decreases noticeably after the first operation during which Xenon atoms are thought to break the surface oxides.
Hence, existence of energetic ions may be considered. Moreover, arc discharges have been observed during the experiments particularly in the higher pressure regime. Arc discharges raises the cathode temperature increasing the electron emission coefficient by one or two order of magnitude [24], and in the process has been observed to modify the surface. Such deformities are detrimental for the formation of self-organization in that they destroy the initial homogeneity of the system.
Chapter 4

Theory and Simulation

Self-Organization is a phenomenon that has been observed across various fields from chemistry to biology to more recently in informatics. It is described as “the spontaneous creation of globally coherent pattern out of local interaction” [11]. Many of the system exhibiting property of self-organization are operating in a state far from thermodynamics equilibrium, and self-organization are thought to be results primarily due to forces tending to equilibration [18]. Occurrence of self-organization has been reported with various types of plasma, and a variety structures and patterns have been observed in different non-equilibrium plasma such as Dielectric Barrier Discharge [19], Barrier Discharges in semiconductor electrode [20] and in plasma with liquid anodes [21]. This has given rise hypothesis that a common underlying mechanism may be at work in these various phenomena. Many different models have been proposed in explaining the observed patterns including the classical model put forward by Alan Turing. Many of the more recent models build on Turing’s seminal work and have proven to be successful in describing the rich patterns observed all across the nature. The more recent models in the field of microplasma are known as drift-diffusion-ionization-Poisson model [19], and are closely related with Turing’s model. They claim, instead of a special mechanism, patterns observed during self-organization originate from non-linear interaction of different plasma components. The model will be explained below. Before describing these models, some relevant phenomenon seen in the discharge have been discussed.
4.1 Excimer Formation

At elevated pressure, Xenon is one of the most efficient excimer sources emitting at 172 nm, with efficiency of 3-5% [17] [22]. High concentration of both high energy electrons, and gas particles are required for excimer production. Excimer production mechanism itself is an important part of self-organization phenomenon. It has been shown that introduction of impurity both decreases excimer emission, while also killing self-organization [13].

The reactions that Xenon undergoes during the excimer formation are studied in reference [17]:

\[
\begin{align*}
Xe^* (1s_5) + 2Xe & \rightarrow Xe_2^* (3\Sigma_u^+) + Xe, & (4.1a) \\
Xe^* (1s_4) + 2Xe & \rightarrow Xe_2^* (0_3^+) + Xe, & (4.1b) \\
Xe^* (0_3^+) + Xe & \rightarrow Xe_2^* (1\Sigma_u^+) + Xe & (4.1c)
\end{align*}
\]

The specimens \(Xe_2^* (1\Sigma_u^+)\) and \(Xe_2^* (3\Sigma_u^+)\) are formed primarily by electron impact excitation as represented in the following equation:

\[
\begin{align*}
e + Xe & \rightarrow Xe^* (1s_4) + e, & (4.2a) \\
e + Xe & \rightarrow Xe^* (1s_5) + e, & (4.2b)
\end{align*}
\]

Another possible reaction involved in excimer radiation through electron impact is as shown in the following reaction:

\[
\begin{align*}
e + Xe & \rightarrow Xe^+ + e + e, & (4.3a) \\
e + Xe^* & \rightarrow Xe^+ + e + e, & (4.3b) \\
e + Xe_2^* & \rightarrow Xe_2^+ + e + e & (4.3c)
\end{align*}
\]
However, the question arises if excimer production is a very important precursor to the observation of self-organization itself. Experiments have shown copper cathode produces comparable excimer intensity with Molybdenum, and aluminum excimer emission at some pressure are three times as high as Molybdenum [12]. However, no self-organization has been observed in CBLD with either Copper or Aluminum cathode. Thus, while excimer emission is an important part of self-organization, it does not always lead to self-organization. Similar arguments have been put forward by Takano [12].

4.2 Filamentation

The development of filaments from the homogeneous discharge is considered as one of the critical stages in the development of self-organization. Filamentation is the process of developing regions of higher concentration of plasma from the state of initial uniform discharge. As discussed earlier filamentation coincides with the entrance into a different phase of current voltage curve. It also correspondingly marks a reversal in the trend of current density in CDVC curve. During experimentation, filamentation were strongly linked with the formation of spots, in the sense that process of filamentation directly led to process of formation of spots. One of the hypotheses in explaining the process of filamentation is identified as the rise in effective secondary emission coefficient coupled along with local temperature effects on the cathode. This would cause increased thermal electron emission, and increased photo emission of electrons. Alternative hypothesis about gas heating was also proposed, however rejected when no high temperature were observed.[12]. Benilov suggests the beginning of filamentation due to uneven cathode surface condition [24]. Using Attractor-Inhibitor model, filamentation can be seen as the process of interaction between attractors and inhibitors, and its evolution can be visualized by modeling the equations governing such systems.
4.3 Negative Differential Resistance

The discharge plasma primarily operates with positive differential conductivity. However, with the onset of filamentation, negative differential conductivity is observed within a small region. This instability is considered as one of the major reasons for the formation of self-organization.

One of the criteria for plasma stability operating in the negative differential region is referred to as the Kaufmann criterion which requires:

\[ \frac{dV}{dI} + R > 0 \]  

(4.4)

where \( \frac{dV}{dI} \) is the differential resistance, and \( R \) is the value of load resistance. It had been noted earlier that the use of additional resistance led to the discovery of the ring structure. As reported by Zhu et. al., for 75 Torr 3 spot situation prior to ring structure occurs at 274 V and 0.083 mA, and ring structure is observed at 276 V and 0.066 mA [9]. The negative differential resistance value for the transition between three spots to ring structure is calculated as 118 KΩ ± 3 KΩ. Hence it is conjectured that due to the usage of ballast resistance of 100 KΩ, slightly smaller than the negative differential resistance, the Kaufmann’s criterion had not been met in previous experiments for the observation of the ring structure. In our experiments, two potentiometers, each with resistance in the range of 0-250 KΩ were used which helped us to readily meet the Kaufmann’s criteria.

4.4 Coulomb Interaction Model

In describing the state of self-organized states, mutual coulomb repulsion between positive space charges and positive surface charges accumulated on the surface of dielectric is identified as the cause, which was reported to be in good agreement with the experimental observations [12][27]. Assuming the charge of each spot to be \( q \), and the linear charge density on the side wall as \( \lambda \), we can express the Hamiltonian of the system for \( n \) spots as:
\[ H = q \sum_{i=1}^{n} \phi_i = \sum_{i} \frac{q\lambda r_0}{4\pi \epsilon} \int_0^{2\pi} \left( r_i^2 + r_0^2 - 2r_ir_0 \cos \theta \right)^{1/2} d\theta + \frac{q^2}{4\pi \epsilon} \sum_{j \neq i} \sum_{j} \frac{1}{|r_i - r_j|} \]  

(4.5)

where, \( r_i \) represents the distance of \( i^{th} \) spot from the center of the hole, which is taken as the origin for the cylindrical coordinate system. \( r_0 \) represents the radius of the hole, which in our case is usually 750 \( \mu \)m, while \( \theta \) represents the angle between the vectors. The spots are expected to arrange themselves in the position corresponding to the least energy. Therefore, the hamiltonian is expected to minimize with respect to \( r_i \). The model is 2D in nature and is primarily focused about the arrangements of the spot themselves.

A python code simulating the system with multiple spots was implemented, which is available in Appendix II. The spots are randomly arranged at the beginning and are allowed to interact electrically and the charge on the wall is defined arbitrarily. The system is allowed to evolve until a stable state is reached. The size of the spots are arbitrarily chosen, and does not affect the final configuration reached. The results were found to be a good match to the experimental finding as seen in fig 4.1.

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The simulation also helped in understanding some of the non-linear characteristics of discharge.
During experiments, more than one configuration for same number of spots were observed, which in turn led to different branches in current-voltage curve. The simulation showed how the final states are depended upon the initial location of the spots. Different initial locations evolved into different final configurations. For instance six spots, depending upon the initial locations of spots, evolved into a regular hexagon, or a regular pentagon with a central spot. Similarly, seven spots evolved into a regular heptagon, or a hexagon with a central spot. Both in experiment and simulation, occurrence of regular hexagon is common compared to regular pentagon structure. On the other hand, a regular hexagon with a central spot is much common than a regular heptagon.

![Figure 4.2: Two alternative configuration of six spots and seven spots configuration seen in simulation and experiment.](image)

While Coulombic interaction does explain the arrangement of spots, this model cannot capture the rich dynamics of plasma, the origin of spots itself, their geometry, and wider range of phenomenon seen in our experiment. Another criticism about the validity of this model comes from lack of explanation about the stability of the spot themselves should they be formed by ensemble of positively charged particles as the resulting repulsion would make the spot geometry highly unstable [25], which cannot exist given high repulsion among the constituent particles. Nevertheless, Coulomb interaction model provides an uncanny match for many of the experimental results.
4.5 Attractor Inhibitor Model

Alan Turing was the earliest proponent of Attractor Inhibitor model. In his seminal paper he uses this simple model to obtain the various patterns in nature. Turing is primarily inspired by morphogenesis during foetal development during which a homogeneous cell mass differentiates into different organs as the zygote develops [23]. Turing uses two different chemicals X and Y, and tries to explain the patterns using their differential diffusion coefficient. The primary equation involved in discretized form are:

\[
\frac{dX_r}{dt} = f(X_r, Y_r) + \mu(X_{r+1} - 2X_r + X_{r-1}) \quad (4.6a)
\]

\[
\frac{dY_r}{dt} = g(X_r, Y_r) + \nu(Y_{r+1} - 2Y_r + Y_{r-1}) \quad (4.6b)
\]

where \(\mu\) and \(\nu\) represent two different diffusion coefficient, \(f(X_r, Y_r)\) and \(g(X_r, Y_r)\) represent the rate of change of X and Y respectively. For modeling in CBLD, these equations were modified by Takano [12] as below:

\[
\frac{dj}{d\tau} = D_j \Delta j - f(j) + \nu \quad (4.7a)
\]

\[
\frac{d\nu}{d\tau} = D_\nu \Delta \nu - pj + q\nu \quad (4.7b)
\]

In the equation, \(j\) and \(\nu\) represent current density and voltage. \(D_j\) and \(D_\nu\) represent the diffusion coefficients, \(f(j)\) represents the charge exchange reactions and \(p\) and \(q\) represent the damping factors of the plasma. Initially a homogeneous discharge is assumed which has homogeneously distributed fluctuations. The function \(f(j)\) is modeled after SNDC (S-Shaped Negative Differential Conductance), which gives rise to Fitz Hugh Nagumo equations. When appropriate coefficients are used, the simulation results in the formation of Turing patterns, similar to those observed in CBLD. More work on this model can be found in Takano’s thesis [12].
4.6 Drift-diffusion-ionization-Poisson Model

This model has been successfully employed to describe different plasma pattern formation. The initial attempt was made by Benilov [24], who used drift, ionization, volume losses and Poisson’s equation. He used 2D Helmholtz equation to model the system:

$$\frac{\partial^2 \Phi}{\partial x^2} + \frac{\partial^2 \Phi}{\partial y^2} + k^2 \Phi = 0$$  \hspace{1cm} (4.8)

The equation was solved for eigenfunction $\Phi$ using homogeneous Neumann boundary condition. For a cylindrical geometry represented by cylindrical coordinates $(r, \theta, z)$ the solution to the equation are:

$$\Phi = J_v(kr)\cos \theta, k = j'_{v,s}/R$$  \hspace{1cm} (4.9)

where $R$ is the radius of the cylinder, $J_v(x)$ is the Bessel function of the first kind of order $v$, $j'_{v,s}$ is a $s^{th}$ zero of the derivative of $J_v(x)$. The results from this model were inconsistent with the experimental observation, primarily in that the solution provided by the model predicted same number of spots in different rings [12]. Pedro et. al. improved and expanded the model using local approximation for electron kinetics and transport [26]. The model posits the existence of a single ion species and allows the system to evolve looking for steady state solution. The equations overruling the discharge are:

$$\nabla \cdot J_i = n_e \alpha \mu_e E - \beta n_e n_i$$  \hspace{1cm} (4.10a)

$$J_i = D_i \nabla n_i - n_i \mu_i \nabla \phi$$  \hspace{1cm} (4.10b)

$$\nabla \cdot J_e = n_e \alpha \mu_e E - \beta n_e n_i$$  \hspace{1cm} (4.10c)

$$J_e = \nabla \cdot D_e \nabla n_e - n_e \mu_e \nabla \phi$$  \hspace{1cm} (4.10d)

$$\epsilon_0 \nabla^2 \phi = -e(n_i - n_e)$$  \hspace{1cm} (4.10e)
Here the parameters $n_i$, $n_e$, $D_i$, $J_i$, $J_e$, $\mu_i$ and $\mu_e$ are number densities, diffusion coefficients, densities of transport fluxes and mobilities of ions and electrons respectively [2]. The coefficient $\alpha$ is Townsend ionization coefficient, and $\beta$ is the coefficient of dissociative recombination. The electric field potential is denoted by $\phi$; $e$ represents the elementary charge and $\epsilon_0$ represents the permittivity of the free space. In order to model the system, a cylindrical discharge vessel is considered of radius $R$ and height $h$. When the origin is located at the center of the cathode, the boundary conditions in cylindrical coordinates $(r, \theta, z)$:

$$
\begin{align*}
  &z = 0 : \frac{\partial n_i}{\partial z} = 0, J_{ez} = -\gamma J_{iz}, \phi = 0; \\
  &z = h : n_i = 0, \frac{\partial n_e}{\partial z} = 0, \phi = U; \\
  &r = R : \frac{\partial n_i}{\partial z} = \frac{\partial n_e}{\partial z} = 0, J_{ir} - J_{er} = 0.
\end{align*}
$$

(4.11)

Here $U$ represents the discharge voltage, $\gamma$ represents secondary emission coefficient which experimentally had been determined as 0.03 [12]. Perhaps to model different metals, different value of $\gamma$ is to be used. The values of different coefficients and results was done by Pedro et. al. and can be found in [2]. An extensive literature giving the recent evidences is present in [24] summarizing the present finding. The coupled equations were solved using COMSOL ® Multiphysics Simulation tool.

Multiple steady solutions for the plasma glow discharges were found using bifurcation theory, and the solutions are categorized as 1D, 2D and 3D solutions. 1D solution consists of plasma operating in abnormal mode and plasma parameters varying only with height; 2D modes involves solution which varies with height and radius, whereas 3D modes varied with height, radius and angle. The simulation aims to qualitatively reproduce the different stable configuration of plasma as has been seen in the experimental results. One of the assumptions used in the modeling was the neutralization of the charge on the wall. Such assumption was made for decreasing computational load. This is the primary cause for some discrepancy in experimental finding and simulation results in that there is very little to no separation between spots and the edge. The issue is being addressed in the upcoming models by the authors. Besides, the model successfully predicted the self-organization in Krypton at higher pressure.
The reason for this has been pointed as the decrease of effective collision cross sections of electrons in Krypton, which is to be compensated by pressure[2]. The model also provides an explanation for a varied number of structures, and it had predicted the ring structure observed later as shown in the fig 4.4. Although no direct quantitative comparison is done, the spots in the model approximately appears to match with the experimental finding that the intensity decays proportionally to the distance from the center.
Figure 4.3: (a) Bifurcation diagram of 3D modes in krypton, $R = h = 0.5$ mm. Solid line: 1D mode. Dashed line: second-generation 3D mode $a_1 b_{10}$. Dotted lines: third-generation 3D mode $a_{10,1} b_{10,1}$. Circles: bifurcation points. Triangles: states for which distributions of current density over the cathode surface are shown in (b). (b) Distributions of current density over the cathode surface for states belonging to the 3D modes shown in a). Bar in Am$^{-2}$. [Image Courtesy of Dr. Pedro Almeida, University of Madeira. ] [2]

Figure 4.4: Comparison between simulations work and experimental finding. [Experiment: W. Zhu and P. Niraula 2014. Modelling: P. G. C. Almeida, M. S. Benilov, and D. F. N. Santos 2013, 2014.]
Chapter 5

Conclusions and Outlook

We are making some strides at understanding the phenomenon of self-organization in CBLD. However, not all of the phenomenon in self organization is completely understood. The focus of research both in terms of experiments and computation had been on the stable mode only, however some of the unstable mode were also seen to exist, and they appeared to show interesting states of self-organization. Since finding unstable states of self-organization would require more sophisticated experimental setting and would be more difficult to study their transient nature, this is considered a task for the future as we will have better understanding of the phenomenon.

Figure 5.1: An unstable mode observed in Krypton at 175 Torr.

Another promising avenue for the research would be study of self-organization in Noble gas mixtures. Although we have not systematically studied the gaseous mixture , the preliminary investigation has shown presence of novel behavior of plasma in the gaseous mixture. The gaseous mixture has been
studied for the industrial purposes for displays. They could also open avenues for application of CBLD. Another unexplored region in CBLD research is exploring states in the transition region. This would require the use of high speed camera. It would also help to settle experimentally the question whether the ring structure is a rotating spot or a stable 2D situation. In addition, we are also interested in exploring the pulsed perturbations in the current existing patterns.

![Figure 5.2: Sprocket and Shuriken like arrangements seen in Steel cathode at 130 Torr.](image)

We were also able to observe the interesting phenomenon in different cathodes. While they followed the similar path as the molybdenum cathode, some cathodes such as Steel at times tended to produce different modes, unlike anything seen before. It remains to be seen if secondary emission coefficient alone is responsible for the rise of such novel phenomena. Also experiments with Argon has failed to yield any self-organization. Experiments with all different parameters should be done to confirm such claim.

Another thing we are interested in to explore is some of the potential application of our experiment. Our future plans includes building an array of holes, and explore if this could potentially be used as a light source. CBLDs are known for the production of excimer sources with the efficiency as high as 5% for Xenon and as high as 2.5% for Argon in excimer production [12]. In addition, the positive CVC allows operation of CBLDs in arrays without a ballast resistance allowing to scale the device [2]. By analyzing the efficiency of such arrayed CBLD, and comparing against the conventional sources of vacuum ultra-violet source, some potential industrial application would be explored.
Chapter 6

Appendix I

Following are the list of the papers that has been published from the results obtained in course of research for this thesis:


Following are the list of the conference papers and paper that were resulted from the research conducted in regards to this thesis:


Chapter 7

Appendix II

The following program simulates the and was written in python 3.3. It also requires numpy module which can be downloaded from www.numpy.org.

```python
import tkinter
from tkinter.messagebox import showerror
import numpy as np
from random import randint
import time

def TestValidLocation(x, y, r):
    if ((x-300)**2+(y-300)**2)**0.5<250-1.5*r:
        return True
    else:
        return False

def CalculateInterSpotForce(locations):
    q = 1
    DistanceAngle = []
    #calculate the distance and the angle

    for i in range(len(locations)):
        IndividualDistanceAngle = []
        for j in range(len(locations)):
            if i!=j:
                x1, x2= locations[i][0], locations[j][0]
                y1, y2= -locations[i][1], -locations[j][1]

                distance = ((y2-y1)**2+(x2-x1)**2)**0.5
                try:
                    #calculating angle for ith particle
                    angle = abs(np.arctan(-(y2-y1)/(x2-x1)))
                except ZeroDivisionError:
                    angle = np.pi/2
                if x1<x2:
                    angle = np.pi-angle
                if y1<y2:
```
angle = −angle  # changing the sign
IndividualDistanceAngle.append([distance, angle])
DistanceAngle.append(IndividualDistanceAngle)

Force = []
for i in range(len(DistanceAngle)):
    Fx = 0
    Fy = 0
    for j in range(len(DistanceAngle[i])):
        theta = DistanceAngle[i][j][1]
        r = DistanceAngle[i][j][0]
        if r!=0:
            Fx += np.cos(theta)*q**2/r**2
            Fy += np.sin(theta)*q**2/r**2
        else:  # if distance is zero to keep the dots moving
            Fx += randint(-10,10)/100
            Fy += randint(-10,10)/100
    Force.append([Fx, Fy])
return Force

def CalculateWallForce(location,q_den):
    q = 1
    steps = 1000
    stepsize = 2*np.pi/steps
    R = 250
    Rs = ((300−location[0])**2+(300−location[1])**2)**0.5
    DeltaX = location[0]−300
    DeltaY = −(location[1]−300)
    if DeltaX != 0:
        theta0 = abs(np.arctan(DeltaY/DeltaX))  # angle made with the center
    else:
        theta0 = np.pi/2

    # accumulators of force
    Fx = 0
    Fy = 0
    for i in range(steps):
        theta = i*2*np.pi/steps
        Denominator = (R**2+Rs**2−2*R*Rs*np.cos(theta))
        if Denominator==0:
            F = 0
        else:
            F = −(q*q_den)/(R**2+Rs**2−2*R*Rs*np.cos(theta)) * stepsize

    if location[0] > 300 and location[1] < 300:
        Quadrant = 1
        Angle = theta0+theta
    elif location[0] < 300 and location[1] < 300:
Quadrant = 2  
Angle = np.pi-theta0+theta

elif location[0]<300 and location[1]>300:  
    Quadrant = 3  
    Angle = np.pi+theta0+theta

else:  
    Quadrant = 4  
    Angle = 2*np.pi-theta0+theta
Fx += F*np.cos(Angle)  
Fy += F*np.sin(Angle)
return [Fx, Fy]

def RunSimulation(NumberSpots, SpotRadius):
    global canvas, SimulateButton, locations, WallCharge, Perturb
    try:
        q_den = float(WallCharge.get())/(2*np.pi)
    except:
        showerror(root, "Invalid entry for charge density. Using default value of 2*pi")
        q_den = 2*np.pi
    SimulateButton.config(state=tkinter.DISABLED)
    Rs = SpotRadius # radius of the spot
    q = 1 # making charge 1
    locations = [] # for storing the location of spots
    for i in range(NumberSpots):
        valid = False
        while (not valid):
            tempLocation = [randint(50, 550), randint(50, 550)]
            valid = TestValidLocation(tempLocation[0], tempLocation[1], Rs)
        locations.append(tempLocation)

    # Drawing the spots
    for i in range(NumberSpots):
        canvas.create_oval(locations[i][0]-Rs, locations[i][1]-Rs, locations[i][0]+Rs, locations[i][1]+Rs, outline="blue", fill="blue", tags="spot%s" %\((i+1)\))

    # Calculating direction of force and performing the animation
    Force = CalculateInterSpotForce(locations)
    ThresholdNoise = 0

    # for adding perturbation
    Perturb = False
    for count in range(500000):
        for i in range(len(Force)):
            WallForce = CalculateWallForce(locations[i], q_den)
Fx = WallForce[0] + Force[i][0]
Fy = WallForce[1] + Force[i][1]
value = 20 / (1.05 ** count)
RateX = int(value * abs(Fx) / (Fx**2 + Fy**2) ** 0.5)
RateY = int(value * abs(Fy) / (Fx**2 + Fy**2) ** 0.5)
if RateX < 1:
    RateX = 1
if RateY < 1:
    RateY = 1
if Fx > ThresholdNoise:
canvas.move("spot%s" %str(i+1), RateX, 0)
locations[i][0] += RateX
elif Fx < -ThresholdNoise:
canvas.move("spot%s" %str(i+1), -RateX, 0)
locations[i][0] -= RateX
if Fy > ThresholdNoise:
canvas.move("spot%s" %str(i+1), 0, -RateY)  # need to check the sign
locations[i][1] -= RateY
elif Fy < -ThresholdNoise:
canvas.move("spot%s" %str(i+1), 0, +RateY)
locations[i][1] += RateY
root.update()

if (Perturb):
    for i in range(len(locations)):
        valueX = randint(-10, 10)
        valueY = randint(-10, 10)
        locations[i][0] += valueX
        locations[i][1] += valueY
        canvas.move("spot%s" %str(i+1), valueX, valueY)  # drawing the spots
Perturb = False
print(locations)

# test if the location is valid and exit until all locations are invalid in when changing both in x and y makes difference
Force = CalculateInterSpotForce(locations)
def BaseSimulation(frame):
global canvas
try:
    canvas.destroy()
    SimulateButton.config(state=tkinter.ACTIVE)
except:
    pass
canvas = tkinter.Canvas(frame, width=600, height=600, bg="#191919")
canvas.pack()
canvas.create_oval(50, 50, 550, 550, outline="white")
def PerturbFunc():
global Perturb
Perturb = True
def SetParameters(frame, sideframe):
global SimulateButton, WallCharge
# to activate it and deactivate it from other functions
# number of spots to be selected by the user
NumberOfSpots = tkinter.Label(frame, text="Number of Spots")
NumberOfSpots.grid(row=0, column=0, padx=10, pady=10)
SpotNum = tkinter.IntVar(frame)
SpotNum.set(1)
NumSpotlist = [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25]
SpotOption = tkinter.OptionMenu(frame, SpotNum, *NumSpotlist)
SpotOption.grid(row=0, column = 1)

# Spot radius
SpotRadiusText = tkinter.Label(frame, text="Radius Size (pixels) :")
SpotRadiusText.grid(row=1, column=0, padx=10, pady=10)
SpotRadius = tkinter.IntVar(frame)
SpotRadius.set(10)
SpotRadiuslist = [5, 10, 15, 20, 25, 30, 35, 40]
SpotRadiusOption = tkinter.OptionMenu(frame, SpotRadius, *SpotRadiuslist)
SpotRadiusOption.grid(row=1, column = 1, padx=10, pady=10)

# Charge Ratio
WallChargeRatioLabel = tkinter.Label(frame, text="Charge Ratio")
WallChargeRatioLabel.grid(row=2, column=0)
WallCharge = tkinter.StringVar()
WallChargeEntry = tkinter.Entry(frame, width=8, textvariable=WallCharge)
WallCharge.set(str(2*np.pi))
WallChargeEntry.grid(row=2, column=1)
tkinter.OptionMenu(frame, SpotNum, *list)

SimulateButton = tkinter.Button(frame, text="Simulate", command=
lambda: RunSimulation(SpotNum.get(), SpotRadius.get()))
SimulateButton.grid(row=3, column=0, padx=10, pady=20)

PerturbButton = tkinter.Button(frame, text="Perturb", command=
lambda: PerturbFunc())
PerturbButton.grid(row=3, column=1, padx=10, pady=20)

ClearButton = tkinter.Button(frame, text="Clear", command=lambda:
BaseSimulation(sideframe))
ClearButton.grid(row=4, column=0, padx=10)
QuitButton = tkinter.Button(frame, text="Quit", command=lambda: root.destroy())
QuitButton.grid(row=4, column=1)

root = tkinter.Tk()
root.title("Simulation")
root.geometry("900x600")
root.resizable(width=False, height=False)
FrameParam = tkinter.Frame(root, width=200, height=600)
FrameParam.grid(row=0, column=1)
FrameDiagram = tkinter.Frame(root, width=600, height=600)
FrameDiagram.grid(row=0, column=0)
SetParameters(FrameParam, FrameDiagram)
BaseSimulation(FrameDiagram)
root.mainloop()
Bibliography


