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The missing modes of self-organization in cathode boundary layer discharge in xenon

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Abstract
Self-organized pattern formation has been previously observed in cathode boundary layer discharges (CBLDs) in high-purity xenon gas at pressures ranging from about 60 Torr to atmospheric pressure. However, certain modes predicted by the COMSOL multiphysics simulation were never observed. In this paper, using the same reactor design, we managed to fine tune the discharge current into regions that were not fully explored before. Two new self-organized patterns were observed, at the verge of the extinguishing of the self-organization. One pattern was a perfect ring that was detached from the dielectric walls. The other pattern was a series elongated spots arranged along a circle. Both patterns were preferably observed at pressures ranging from 60 to 120 Torr. The observation of these patterns may open up new discussions to the self-organized pattern formation in CBLD in xenon.

Keywords: self-organized, pattern, xenon, direct current, cathode boundary layer discharge, plasma spots

1. Introduction
Self-organized pattern formation in direct current glow discharges in xenon was first reported by Schoenbach et al [1, 2]. The plasma device involved in their study was comprised of a flat cathode and a ring-shaped anode that were separated by a thin layer of a perforated dielectric spacer. The inner diameter of the anode ring (as well as the diameter of the opening on the dielectric spacer) varied from 0.75 to 3.5 mm and the thickness of the dielectric spacer ranged between 100 and 250 µm. Different from the micro-hollow cathode discharge (MHCD) that was reported earlier by the same group [3], the expansion of the plasma on the cathode surface is limited by the walls of the opening on the dielectric spacer. This type of discharge was then referred to as the cathode boundary layer discharge (CBLD). Self-organized plasma patterns were observed when lowering the discharge current to certain values, which was accompanied by an abrupt discharge voltage drop. These patterns form in CBLD in xenon regardless of whether there was a tiny dip at the center of the cathode surface, and the plasma spots were strong vacuum ultraviolet (VUV) emitters. Takano et al [4] modified the device to a plane-to-plane structure while maintaining the circular opening on the dielectric spacer, and observed similar self-organized plasma patterns in xenon. Zhu et al [5] expanded the device from a single circular hole to multiple holes sharing the same anode and cathode layers, and to a long shallow trench (1.5 cm long, 0.25 mm wide and 0.25 mm deep), where plasma patterns were consistently observed in xenon. It was suggested in these publications that the plasma first forms filaments, perhaps due to an increase in the effective secondary emission coefficient caused by high energy photons (with energies higher than the work function) striking specific areas on the cathode. Then this filamentary plasma forms patterns because of Coulomb interactions between the positively charged cathode fall channels and positive charges on the surface of the surrounding dielectric spacer. Benilov [6, 7], however, argued that basic mechanisms of glow discharge (i.e. drift and diffusion of the ions and the electrons, volume ionization and recombination and secondary electron emission) was sufficient to explain the pattern formation in dc glow discharges, and suggested that the patterns were simply multiple solutions existing at the same discharge current in the theory of glow discharges. The simplest case of these solutions describes states with a uniform distribution of current over the electrode surface and is referred to as the 1D mode (solution). Axially symmetric solutions correspond to the diffusion of the charged particles (ions and electrons) to the (absorbing)
walls and are referred to as the 2D mode (solution). 3D mode (solution) carries an arbitrary azimuthal period (2π, or π, or 2π/3, or π/2, etc). A detailed description of the mathematical formulation of the theory of multiple solutions and explanation of the different modes can be found in a review paper by Benilov [8] in this cluster issue. Some cases predicted by the modeling corresponded well to experimental observations, while others seemed impossible to be realized physically (e.g. a ring-shaped plasma spot located in the interior of the cathode, see figure 1(c) in [9]). In this short communication, we present for the first time experimental proof of the formation of the ring-shaped plasma spot, as well as one other intermediate mode observed during the transition from well-defined plasma spots to the ring plasma mode.

2. Experimental setup

Molybdenum foil (Alfa Aesar, thickness: 250 µm and purity: 99.95%) was used as both the cathode and the anode material. A circular hole of 750 µm in diameter was drilled through the anode manually via a stainless steel drill bit on a mini mechanical drill press. Quartz disk (thickness: 250 µm) with predrilled circular hole (Φ = 750 µm) was used as the dielectric spacer. A circular molybdenum disk (Φ = 4.76 mm) was used as cathode layer. All layers were cleaned with acetone and methanol in an ultrasonic bath for 5 min, rinsed with deionized water to remove residual solution, then assembled with Torr Seal® and annealed in air in a box oven at 120 °C for half an hour. The finished device was then carefully installed onto a grounded aluminum sample holder inside a stainless steel vacuum chamber, which was pumped down to a base pressure of ~0.4 mTorr. Research grade xenon (99.999%) was introduced into the chamber to the desired pressure and kept static until the next pump down and refill cycle. A copper wire was used to connect the cathode to an electrical feedthrough on the chamber, which was further connected to a current limiting dc power supply (Glassman®) via a ballast resistor (R = 100 kΩ) and a current monitoring resistor (r = 1 kΩ). To better facilitate the fine tuning of the discharge current, two high-voltage potentiometers (R = 0–200 kΩ) were also connected in series in the circuit. Discharge voltage was taken via a multimeter that can measure up to 1500 VDC. Discharge current was converted from the voltage measured across the current monitoring resistor. An over voltage (current limited to approximately 1 mA) was provided for the initial ignition of the device. Current was then gradually reduced until self-organized patterns appear. Further current reduction was achieved by adjusting the potentiometer built into the power supply and the two high-voltage potentiometers connected in the circuit.

A Panasonic GP-KR222 charge-coupled device (CCD) camera together with a telescope lens was used to record visible video (25 frames per second) of the discharges through a quartz viewport on the vacuum chamber. Snapshots of the discharges were digitally recorded via a Hauppauge® WinTV® video box. A schematic diagram of the experimental setup as well as a close-up view of the plasma device is shown in figure 1. Image analysis was carried out with Image J (v1.47, National Institute of Health).

3. Results and discussions

In a typical current–voltage curve of CBLD in xenon, the current region within which self-organizations occur is pressure dependent (e.g. figure 12 in [5]). The general trend is that at higher pressures (p ≥ 175 Torr), the self-organization region spans a wide current range (>2 mA) with
Figure 2. The development of plasma spots in CBLD at a xenon pressure of 75 Torr. Voltages and currents are noted under each image. White circles designate the boundaries of the circular opening. Image (30) corresponds to a steady ring plasma mode.

high numbers (many layers) of plasma spots in small sizes. While at lower pressures \((p \leq 125 \text{ Torr})\), the self-organization region appears in a very narrow current range \((<0.5 \text{ mA})\) with fewer numbers of plasma spots in bigger sizes that are well separated. The pressures used in this study range from 60 to 120 Torr, which required very careful adjustment of currents in order to obtain different plasma modes. In previous studies of self-organizations in CBLD in xenon, current was almost always adjusted through the potentiometer built into the power supply. The extinguishing of the plasma patterns was usually the result of further reduction in the current when the minimum number of plasma spots was reached. The minimum number of plasma spots that can be sustained in CBLD in xenon is pressure as well as hole diameter dependent (see, e.g., figures 4 and 5 in [4]). The two high-voltage potentiometers connected in series in the electrical circuit made it possible for us to manipulate current at microampere scale in this situation.

3.1. The ring plasma mode

Figure 2 shows the development of the plasma spots at a xenon pressure of 75 Torr. White circles are superimposed on these images to designate the boundaries of the circular opening on the sample. The plasma first filled the circular opening
completely at a current of approximately 1 mA (picture not shown in figure 2). Reducing current led to the reduction of the luminous area gradually until the plasma completely stripped away from the walls (images (1)–(10)). Starting at a current of 0.198 mA, plasma patterns started to develop. There were two distinctive modes of patterns observed: (a) the diffused mode (images (11)–(24)) and (b) the confined mode (images (25)–(29)). In the diffused mode, plasma spots were not circular and the inter-spot space was first occupied by plasma with lower luminescence. When current was decreased, the number of plasma spots increased while the area of each spot decreased, and the inter-spot luminescence diminished. When the maximum number of plasma spots was reached (in this case, 6 spots as shown in image (17) of figure 2), further reduction in current led to the decrease in the spot size only (images (17)–(24)) until it bridged into the confined mode with six circular spots (image (25) of figure 2). In the confined mode, plasma spots remained circular and decreased in number when current was decreased, until the minimum number of spots (in this case, 3 spots as shown in image (29)) was reached. Further reduction in current, however, led to a ‘ring plasma’ as illustrated in image (30) of figure 2. The recorded current and voltage of this ring plasma were 0.066 mA and 276 V, respectively. Further analysis of image (30) revealed that the inner and outer diameters of the ring were approximately 220 µm and 300 µm. The luminescence of the ring was weaker than that from the spots in the previous images.

The current–voltage and current–density–voltage curves are plotted in figure 3, with data points corresponding to transition images in figure 2 annotated. A program developed in-house was used to evaluate the luminous area pixel by pixel in each picture. Pixels with signal intensity above a preset threshold (average of background taken far away from the luminous area) were considered contributing to the current conduction and used for the current density calculation. The curves look very similar to what has been reported by Takano et al [4] with the exception of the data points from the three spots and the ring plasma mode.

In the current–voltage curve, a small slope is observed in the abnormal glow mode, followed by a much bigger slope in the diffused mode of self-organization. In the confined mode of self-organization, a nearly zero slope is observed, as the voltage barely changed from one self-organization pattern to another. These are also clearly observed in the current–density–voltage curve. The reversal of the current density appears at the transition from abnormal glow mode to the diffused mode of self-organization. The highest current density corresponds to the confined self-organization with the fewest number of spots (image (29) in figure 2). The current density of the ring plasma is, however, much lower, reaching a value close to that of the onset of the confined mode of self-organization. This is due to the drop of the plasma current and the increase in the luminous area in the ring plasma.

It has to be noted that the ring plasma carried considerably lower current compared with what was required to sustain the lowest number of plasma spots. It was very likely one of the 2D modes predicted by Almeida et al [9]. This ring plasma mode was consistently observed across different samples from 60 to 120 Torr in xenon at the verge of the extinguishing of the plasma spots. A few further ring plasma pictures at different pressures are shown in figure 4. The rings observed from 60 Torr to 75 Torr (figures 4(a)–(c)) are very similar in their
appearance (inner and outer diameters) and overall currents. The two rings observed at 100 Torr (figures 4(d)–(e)) were taken in different samples. Image (d) corresponds to a fresh refill of xenon in the vacuum chamber with a base pressure of ∼0.4 mTorr while figure 4(e) corresponds to an approximate 0.5% impurity (roughly estimated from the pressure change due to air purposely leaked into the chamber) in the vacuum chamber. The green shift of the ring’s color is likely due to the nitrogen in the air leaked into the chamber. Figures 4(f)–(h) were taken from three different samples, where the rings did not appear to be very symmetrical. Image (h) again corresponds to an approximate 0.5% impurity in the vacuum chamber. The size of these three rings also appeared to be bigger, which will be explained in detail in the next section.

It is also worth noting that in the situations of images (e) and (h), self-organized plasma spots were also observed prior to the formation of the ring plasma, which contradicts the 99.999% xenon purity requirement for the generation of self-organization in CBLD as reported in [4]. We believe this was again due to the addition of the two high-voltage potentiometers in the electrical circuit. Unfortunately, we cannot precisely control the amount of impurity in the vacuum chamber at the current stage. More experiments will be carried out to further investigate the influence of impurity.

3.2. The segmented plasma spots

One immediate question is that whether this ring plasma mode is a steady state or is it simply the rotation of plasma spots at such a high speed that the CCD camera is not able to catch at a refreshing rate of 25 fps. This question led to the identification of the segmented plasma spots. Here we use the development of the ring plasma at 100 Torr as an example. When five plasma spots (minimum number of spots before the plasma extinguishes) were obtained, current was very carefully lowered by adjusting the high-voltage potentiometers. Interestingly, the plasma spots became elongated (figures 5(b)–(e)) until the head of one segmented plasma spot touched the tail of an adjacent segmented plasma spot, forming a ring plasma with apparent nodes (figure 5(f)). The nodes seemed to have disappeared in figure 5(g) and the ring plasma clasped into a smaller ring (figure 5(h)). The transition from (a) to (g) was reversible while the transition from (g) to (h) was unidirectional. Increasing current from (h) led to the extinguishing of the ring plasma. The currents corresponding to figures 5(a)–(g) were very similar with the difference between (a) and (g) approximately 7 µA. While the current difference between (g) and (h) was approximately 28 µA. Increasing current from (a) also led to the expansion of the plasma spots. But the expansion was more symmetrical within each spot and the pattern eventually transitioned to more plasma spots. It has to be noted that depending on the surface condition and the resistance included in the electrical circuit (due to the potentiometer) at the beginning of the experiment, the minimum number of plasma spots that could be sustained in the same sample at the same xenon pressure might slightly differ (e.g. at 100 Torr, the minimum number of plasma spots could be 4 or 5). Nevertheless, these segmented plasma spots were consistently observed across different samples and at different xenon pressures (with different minimum number of plasma spots as the starting point). We believe that they are transient 3D modes. Interestingly, these segmented plasma spots tended to self-regulate to circular plasma spots if left alone (i.e. the current self-regulated until it reached the circular spots situation). This self-adjustment process usually took a few seconds and was clearly observable. Further experiments will be carried out to better understand this phenomenon.

Figure 4. The ring plasma obtained at different pressures from 60 to 120 Torr. Images (e) and (h) correspond to 0.5% air impurity in the vacuum chamber.
3.3. Other missing modes

With the assistance of the two high-voltage potentiometers, we were also able to obtain a single plasma spot at the center of the circular opening at a xenon pressure of 50–66 Torr. This was rather difficult and was previously achieved only through reducing the diameter of the circular opening of the sample to $\sim 300 \mu m$ (see figure 5(d) in [4]). Also observed was a target shape plasma pattern, with a central spot and a ring plasma located at the interior of the circular opening. These two situations are shown in figure 6. The target shape plasma pattern (figure 6(b)) seemed to be at steady state, yet a small current increase (\(\sim 5 \mu A\)) led to the development of clear nodes in the ring plasma (figure 6(c)). Further current increase resulted in the alternate six plasma spot mode—in this mode, instead of the six plasma spots distributed at the corners of a hexagon, five of them were located at the corners of a pentagon and one was located at the center of the circular opening. This alternate six plasma spot mode was less stable than the hexagon situation. Both of these two plasma patterns were believed to belong to the 3D plasma modes.

4. Conclusions

By adding two high-voltage potentiometers in the CBLD electrical circuit, the plasma current was manipulated at microampere level, which led to the development of the ring plasma and the segmented plasma patterns. We believe that the ring plasma was one of the 2D plasma modes whereas the segmented plasma patterns, the single plasma spot and the target shape plasma pattern, were 3D plasma modes. Self-organized plasma patterns and the ring plasma were also observed in CBLD in xenon with $\sim 0.5\%$ air impurity. It may be possible in the future to purposely add trace amounts of nitrogen while maintaining the plasma patterns to further probe plasma kinetics and estimate gas temperatures through the analysis of the optical emission spectra of the second positive system of nitrogen.

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