

# Chapter 1

## Introduction

### 1.1 Overview

#### Motivation

Plasmas, commonly called the fourth state of matter, are a gas where a significant fraction of the neutral atoms or molecules have been split into pairs of electrons and positive ions. Initially, a curiosity of the laboratory, they have become a critical part of every day life. The electrically charged nature of plasmas makes them a practical means by which to convert electrical energy into light, chemical reactions, kinetic energy, or even nuclear reactions. From an applications perspective, they are indispensable in lighting, semiconductor manufacturing, plastic processing, and space propulsion. On a more broad scale, virtually all observable light in the universe is the result of a plasma in some form or another.

Some exceptions aside, only three things are required to create a plasma: a

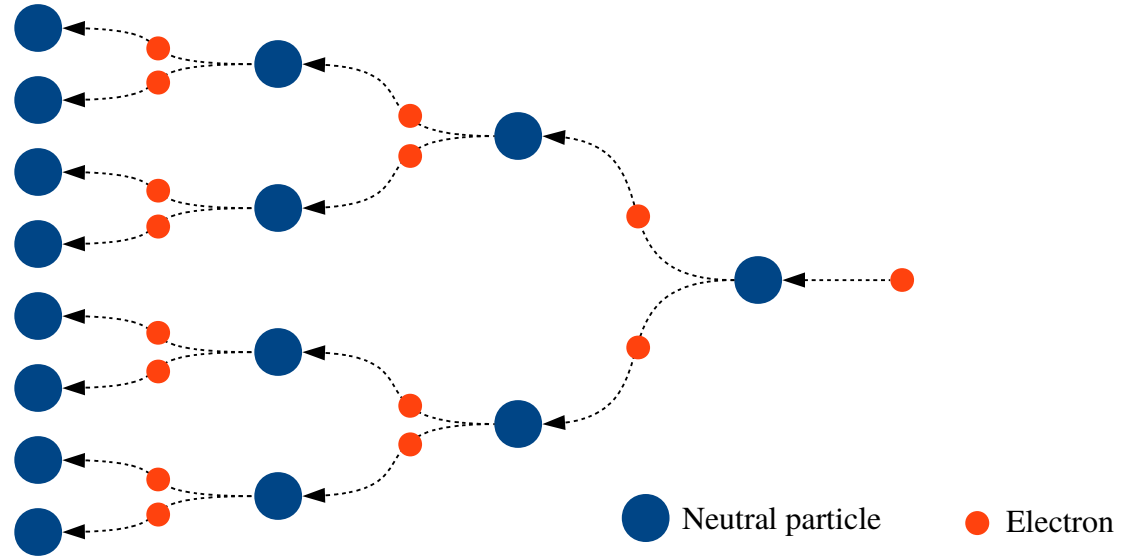


Figure 1.1: A simplified depiction of the avalanche breakdown process in a gas.

gas, an energy source, and a means of transferring the energy to the gas. In man-made applications, the energy source is typically electricity, and the simplest transfer mechanism are two electrodes placed on either side of the gas. This results in an electric field in the gap between the electrodes, as seen in figure 1.1. The field accelerates a single seed electron in the gas (often created by background cosmic radiation) until it collides with a neutral particle. The electron, having acquired a fair amount of energy, knocks a second electron loose from the particle, leaving behind a relatively heavy and immobile ion. Subsequently, both the first and second electron are now accelerated by the electric field. Again, they collide with two more neutral atoms, creating two new electrons. As long as the electric field persists, the number of electron and ion pairs increases exponentially. This process is generally referred to as an avalanche.

Despite this relatively simple recipe, the physical characteristics can vary greatly depending on what gas is used, what pressure it is at, what voltage is used, whether the electricity is applied constantly or varied over time, what kind of electrodes are used, etc. As a result, man-made plasmas are generally produced under very specific conditions. For example, a plasma etcher used in semiconductor manufacturing may need to operate at pressures that are one-thousandth of atmospheric pressure with ultra-pure (99.9999%) gases.

Plasmas like those which occur in plasma etchers, feature ions and neutral gas particles with temperatures that are below 1,000 K, or roughly 1,340° F. Though this temperature is relatively high compared to room temperature, it is well below the temperature of the electrons which may be in excess of 20,000 K. Plasmas which exhibit this disparity in temperatures are often called nonequilibrium or “low-temperature” plasmas.

Conversely, there exists another class of plasmas where the electrons, ions, and neutral particles are all at the same temperature. These are called thermal plasmas. Generally speaking, the closer the gas pressure is to atmosphere, the more thermal a plasma is. Additionally, the temperatures in a thermal plasma can be quite high and, in many cases, can easily melt most metals. For example, the arc of an arc welder is a thermal plasma. Similar high temperature plasmas, have a number of other applications which include a variety of high-intensity lamps, metal cutting, and surface coating.

There are, however, a number of applications which would benefit from operation at higher pressures, but with low-temperature ions and neutrals so as to

avoid heat damage. This has spurred a substantial amount of research on nonequilibrium atmospheric-pressure plasmas (APPS) in recent years. Ideally, such a plasma could be generated at or near atmospheric pressure with hot electrons, but minimal heating of surrounding gas. Though this field of research is still relatively young, it has produced a variety of new plasmas and capabilities. One of the more ubiquitous examples is use of plasmas to process the surface of plastics so that ink can adhere. Separately, nonequilibrium APPS are the technology which drives plasma televisions.

As mentioned, these applications promise to be the first of many for such plasmas. More recently, there have been innovative proposals to use these plasmas in water purification, wound sterilization, improved combustion engines, nanoparticle production, and more. However, each situation has its own challenges when it comes to the design and development of a plasma source, particularly at these elevated pressures. Particularly problematic is the tendency of APPS to develop instabilities which can cause them to rapidly transition to thermal plasmas in a matter of nanoseconds.

There exist a few ways of getting around these instabilities. One example is the dielectric-barrier discharge which passively regulates the amount of power which can be deposited into the plasma. Another example includes split-ring resonators which use natural feedback mechanisms to damp out potential instabilities. The technique considered here, referred to as the repetitively-pulsed nanosecond discharge, or RPND, uses high voltage pulses which are so short that the instability does not have time to develop. The RPND is a nonequilibrium

plasma which can operate at pressures ranging from approximately  $10^{-3}$ –10 atmospheres. At atmospheric pressure the RPND can produce a uniform plasma in volumes on the order of 10 mL. As the pressure is reduced, the plasma volume can reach the order of liters.

The importance of large-area, uniform, high-pressure plasmas such as the RPND was highlighted in the National Academies' most recent decadal survey of plasma science [1]. However, there is still much that is not known about such plasmas. From the same survey, it is said that "the full promise of APPS will be known only if they can be understood and managed based on fundamental scientific principles at two extremes—the nanoscopic kinetic level, where selective chemistry occurs, and the global stability level." It is this challenge, specifically the investigation of the nanoscopic kinetic level, which drives the research presented here.

## History

Historically, the study of low-temperature APPS has been almost indistinguishable from the study of plasmas as a whole. However, this was not necessarily a matter of reasoned choice. Plasma generation at atmospheric-pressure obviates the need for an effective vacuum pump. Additionally, prior to the creation of large battery banks, early sources of electrical energy had relatively small capacities. This precluded the generation of thermal atmospheric plasmas which required large amounts of energy.

Indeed, the requirements for a low-temperature APP are sufficiently rudimen-

tary that the first man-made one (and likely the first man-made plasma), was probably a spark generated by rubbing fur against amber. This is commonly attributed to Thales of Milêtus from around 600 B.C. Following Thales, electrical sparks came to intrigue many scientists including Gottfried Liebniz, Benjamin Franklin, and Charles Wheatstone. By the mid-1800s, Plücker, Geißler, and Hittorf began some of the first work on low-pressure plasmas though it was Crookes who would later identify plasma as a separate state of matter. Later, J.J. Thomson's discovery of the electron and discretized charge in 1897 marked the beginning of modern plasma research.

By this time, the necessary tools and techniques existed to create steady plasmas in pure, rarified gases. The behaviors of which were dominated by the motion and interaction of the charged electrons and ions. Critically, the effects of the neutral particles were negligible, thus isolating the electrical properties of the plasma. These carefully controlled systems were ideal for basic studies of plasma behavior and were used to great effect by individuals such as Irving Langmuir and Lewi Tonks. In fact, many modern concepts in plasma physics can be traced back to their work.

In contrast, the pulsed APPS, characteristic of the earliest man-made plasmas, were easy to create, but notoriously difficult to work with. It could take them only a few nanoseconds to form, and less than a millisecond to decay away. For many years, there were simply no instruments capable of taking measurements this quickly. Furthermore, the neutral particles which were of no consequence in the low-pressure plasmas, could not be ignored. The neutral particles were

present in such quantities that they could confound or obscure otherwise simple measurements.

As a consequence, there is still a great deal that is not known about about pulsed APPS, particularly lightning, streamers, and a type of plasma which Thomson referred to as a “luminous front.” By the 1970s, this latter plasma had come to be called the fast ionization wave, or FIW. It was generated by a single voltage pulse lasting around 100 nanoseconds and peaking at 10s or 100s of kilovolts. For the right pressure and gas, the FIW could fill volumes of nearly 40 L with a relatively uniform plasma, but with little heating of the gas.

These properties were attractive for a number of uses, but the FIW faced a number of implementation-related challenges. The switches used to trigger the FIW could only operate up to 100 times each second. Unfortunately, the lifetime of a plasma at elevated pressures is relatively short, and the plasma generated by the FIW would decay away quickly after each pulse. This meant that the FIW-generated plasma had a relatively low duty cycle; the ratio of the time the plasma spends on to the time it spends off. This was disadvantageous for plasma-processing applications where low duty cycles are equivalent to long processing times. The low duty cycle also necessitated so-called preionization of the gas with UV lamps or a secondary plasma generator, adding to the cost and complexity of the system. Finally, the pulse generators used for FIWs were not considered reliable enough for long operational lifetimes.

Recent advances in solid-state switching technology has largely solved these issues. At present, switches exist which can reliably operate 100,000 times a sec-

ond; sufficiently fast that the plasma duty cycle approaches 100%. This has the additional benefit of obviating the need for a preionization stage, as a sufficient number of electrons persist between pulses. The discharge produced by the use of these new switches is what we refer to as the RPND.

## Questions

The large pedigree of pulsed plasma research belies the fact that they are still not well-understood. This remains especially true for RPNDs which present significant experimental challenges. A major component of this has to do with the time scales associated with the RPND. The formation of a RPND often requires no more than 10-20 nanoseconds. Very sensitive equipment is required in order to measure changes which occur during this period. Unfortunately, such equipment is particularly susceptible to the broadband electronic noise generated by the fast pulses. There is a plethora of other problems that can be traced back to topic of timing. For example, things like the length and insulators of detector cables can introduce substantial delays, and must be considered in order to synchronize different measurements.

Consequently, the majority of RPND studies focus on measurements after the discharge has occurred, when changes happen at a much slower rate. A great deal of information is available for this period of time, including chemical compositions, atomic densities, electron densities, gas temperatures, and more. While undoubtedly important, these measurements provide limited insight on what is happening *while* the plasma is forming. It is natural, then, to ask, what are the



RPND plasma properties during formation?

Additionally, most studies have used a limited range of gases: oxygen, nitrogen, air, hydrogen, or some mixture thereof. The choice of these gases is deliberate and reflects specific applications in combustion and aerospace. However, the use of rare gases (such as helium) and rare gas mixtures has become popular because they provide for a wider range of stable operating conditions. Notably, it has been found that unique internal structures of rare gases can produce very different discharges. Given this, there is the question of how rare gas RPNDs compare to more conventional ones.

Finally, the persistence of the plasma between pulses makes the development of a RPND very different from a FIW. One reason for this is the large number of electrons that remain between pulses in the RPND. While these seed electrons are necessary for the discharge to develop, in too large a number they can shield out the applied pulse. This can have the detrimental effect of reducing the final plasma density. These competing effects have to be balanced in order to obtain an optimal result, and what is considered optimal can vary depending on the application. Therefore, one must ask how the properties of a RPND compare to a similar FIW and how they vary depending on the operating conditions.

## **Approach**

The dissertation presented here represents my efforts to either answer or provide a foundation to answer these questions. In order to develop the appropriate context for this work, the next section will be a comprehensive review of the RPND

literature. It begins with the first reported pulsed APPS and concludes with contemporary studies.

The following two chapters set the basis for the experimental and numerical studies. Chapter ?? presents the theory necessary to understand RPNDs from basic gaseous discharges to cathode-directed streamers. Subsequently, chapter ?? describes the design of the helium RPND discharge apparatus used for the experimental studies and as the basis for the simulations. Also included in this chapter are several measurements of the basic discharge properties.

Chapters ?? through ?? provide more detailed measurements and analysis of the RPND dynamics. In chapter ??, the measurements of the helium metastables in a RPND are presented and analyzed as a function of pressure and axial location in the discharge apparatus. Chapter ?? presents and analyzes similar measurements of the spontaneous plasma emissions. Finally, chapter ?? discusses the development of a global model for a helium plasma and its use with the experimental data to infer the plasma properties of the RPND. The dissertation concludes with a summary of the results and suggestions for further avenues of research.

## 1.2 Literature Review

RPNDs are only a recent invention which resulted from advances in fast-switching semiconductors. However, the physics of their formation is related to a much more broad category of plasmas which includes lightning, streamers, and even some transient phenomena in DC glows [2]. These plasmas are unique in that

their spatial structure develops at speeds much faster than can be accounted for by the conventional Townsend mechanism. Loeb refers to this phenomena as “ionizing waves of potential gradient.”<sup>1</sup>

## Early History of Pulsed Discharges

In 1835 (as reported by Thomson [3]), Charles Wheatstone attempted to measure what he thought to be the speed of electricity between two electrodes, connected by a vacuum tube, six feet in length [4]. It is now known that he was actually measuring the speed with which a plasma formed between the two electrodes. He accomplished this by the use of a rotating mirror which allowed him to see images of two sections of the tube, slightly displaced. The displacement between the images was proportional to the speed with which the plasma traveled between them. Wheatstone estimated this speed to be at least  $8 \times 10^7$  cm/s.

Interestingly, von Zahn later noted that this was *not* the speed of the emitting particles [5]. That is, even though the visible light crossed the electrode gap at a high speed, the particles did not.

Later, Thomson revisited this work with an improved apparatus [3]. This included a tube that was now 15 m in length and five mm in diameter, as seen in figure 1.2. Also using the rotating mirror apparatus, Thomson was able to greatly improve on the estimates of Wheatstone. He estimated that the so-called “luminous front” had a speed that was more than  $1.5 \times 10^{10}$  cm/s, or in excess of

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<sup>1</sup>It should be noted that the phrase wave does not indicate any kind of periodic motion or spatial arrangement. Simply put, it describes a boundary which separates ionized and unionized gas which travels from one electrode to another.

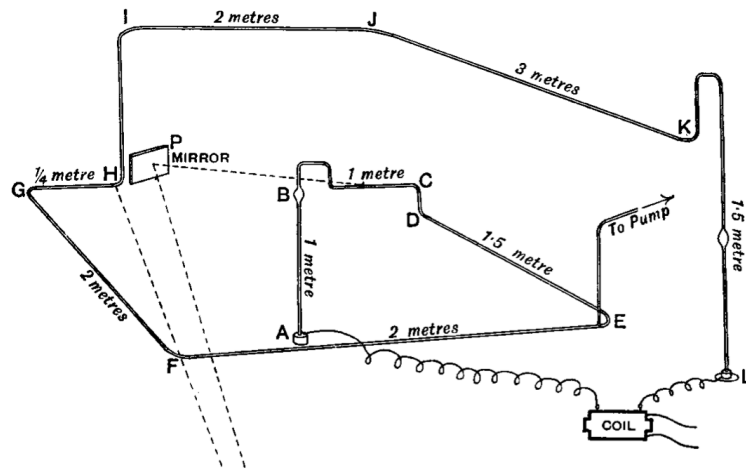


Figure 1.2: A sketch of J.J. Thomson's early experiments on pulsed plasmas in long vacuum tubes.

half of the speed of light. Furthermore, Thomson determined that the luminous front always appeared to travel from the positively pulsed electrode (anode) to the ground electrode (cathode).

The study of these luminous fronts was revisited by several researchers in the wake of Thomson [6–8], but their attempts to duplicate the measured speeds were met with varied success. In 1930, Beams confirmed definitively those of Thomson. He also discovered that the front always traveled from the electrode with the highest absolute potential, to the lowest one. In other words, it could be anode *or* cathode directed, depending on the magnitude of their potentials relative to ground. Beams hypothesized that the rapid motion of the front was a result of a self-propagating region of high space charge, quote:

In the neighborhood of the electrode . . . the field is very high and

intense ionization should take place. This ionization due to the large difference in mobilities of positive ions, negative ions and electrons respectively should result in the establishment of a space charge. This space charge, once formed near the high potential electrode  $Q$  must move down the tube regardless of the polarity of the applied potential because of the changes it produces in the field near its edges.

At about the same time, Schonland and Collens reported on their observations of lightning [9]. Though the general structure and length scale of lightning is substantially different from the luminous fronts observed by Beams and Thomson, the two phenomena would later prove to be very similar. In their work Schonland and Collens noted that lightning would usually occur in a two-step process. Based on the images they obtained, they suggested that the leader was generated by a relatively small “dart” with a mean vertical velocity of  $7.2 \times 10^8$  cm/s. The dart moved in a random manner, changing directions at random intervals, but always moving toward the ground.

The second step began when this dart reached the ground. Once there, a bright return stroke would occur along the same path that the leader had traced out. In contrast to the leader stroke, the return stroke had a velocity of  $5 \times 10^9$  cm/s. Schonland and Collens hesitantly attributed the leader stroke to an extended electron avalanche, and the return stroke to thermal ionization along the conductive path generated by the dart. However, calculations by Cravath and Loeb showed that the speeds of the proposed avalanche was inconsistent with the fields at the head of a lightning stroke [10]. Instead, they suggested that the dart was actually

a moving region of space charge which locally accelerated electrons to ionizing energies. This was similar to the mechanism earlier proposed by Beams.

### **The Streamer Model**

It was long known that sparks in air were similar to lightning. Advances in technology during the 1930's led to experiments which reinforced this similarity. In response to the measurements of Schonland and Collens; Snoddy, Beams, and Dietrich studied the breakdown of gas in a long tube with both positive and negative applied potentials [11]. Using an oscillograph, they observed both the leader and a return stroke in both cases. However, the propagation of the plasma wave toward the cathode required a source electrons ahead of the wave. The authors proposed that photoionization might provide these necessary electrons.

Around the same time, Flegler and Raether had come to a similar conclusion regarding the importance of photoionization. This led them to develop a more thorough theoretical model for these waves [12] which came to be known as the streamer theory. This was followed by a similar treatment by Loeb and Meek [13–15]. The streamer theory divided the initial plasma formation into two steps. In the first step, an electron avalanche is initiated between two electrodes. The avalanche travels toward the anode and leaves behind a region of positive space charge. In the second step, the return stroke begins at the anode and travels along the conductive path generated by the initial avalanche toward the cathode.

The streamer model proved relatively successful in describing the development of sparks and lightning. Theoretical estimates of the speed matched the

velocity measurements that were acquired with photographs and oscillographs. Additionally, the theory was able to account for the halting manner in which lightning was formed as well as the constriction in space.

Following the initial work of Flegler, Raether, Loeb, and Meek, a number of researchers began to explore the boundary between the Townsend mechanism and the streamer mechanism. Most notable was Fisher and Bederson's work in 1951 [16], which was later extended to nitrogen [17] and argon [18]. These studies suggested that the streamer theory was incomplete. Furthermore, the reliance of the streamer theory on photoionization would later prove very contentious [19]. Finally, there was a whole class of discharges that it did not readily explain.

## **Diffuse Streamers**

Per Chalmers [20], Rogowski and Buss [21,22] observed a fast, diffuse, glow discharge immediately prior to the filaments of a streamer discharge. Allibone and Meek, noted similar diffuse discharges in air based on oscillographs and photographs [23–25]. However, the Boys apparatus which was employed in these studies (an ancestor to the modern streak camera) was unable to capture the evolution of the diffuse glow, given its large spatial extent.

This was first noted by Allibone who attempted to use Lichtenberg figures<sup>2</sup> to definitively capture this diffuse glow [26]. Later, Saxe and Meek used the recently invented photomultiplier tube to record the evolution of the light emis-

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<sup>2</sup>Such figures directly exposed photographic emulsions to the electrical discharge. The developed image was a time-integrated representation of the discharge.

sions in the brief, diffuse glow [27] as a function of space. Both studies agreed in the existence of the diffuse glow, despite some disagreement on the nature of its geometry and propagation.

By 1968 (according to Kunhardt and Byszewski [28]), Stankevich and Kalinin had provided the most firm evidence yet of a diffuse discharge in a dense gas [29]. This was later confirmed by experiments with a pulsed nanosecond discharge by Mesyats, Bychkov, and Kremnev [30]. In their analysis, they noted that photoionization could not play a role in such short-lived discharges and thus there was a need for extensions to the streamer model.

In addition to the diffuse discharge, Stankevich and Kalinin also noted the detection of x-rays with each pulse. This suggested the presence of high-energy electrons impinging on the surface of the electrodes, despite the high collisionality of the dense gas. Not only that, but the electron energies could even exceed what would be expected from the vacuum electric fields [31]. The eventual conclusion was that the electric field associated with the space charge at the head of the streamer produced very energetic electrons which deposited their energy far from the streamer tip [28, 32], allowing the streamer to spread out beyond the diffusive region of the electrons.

It was based on the studies of the fast electrons in these discharges that Mesyats, Bychkov, and Kremnev proposed the use of a fast electron beam for pumping high-pressure gas lasers. Similar work was conducted simultaneously by Fenstermacher et al. [33]. Later, Hunter [34], and Koval'chuk and Mesyats [35] proposed that such discharges be used for fast-closing switches. The need for



a homogeneous plasmas in these applications prompted Palmer [36], and Levatter and Lin [37] to investigate the necessary conditions for uniformity. They concluded that the primary requirement for uniformity was a threshold value of fractional ionization.

### **Repetitively-Pulsed Nanosecond Discharges**

The type of discharge originally studied by Babich, Loika, and Tarasova came to be known as the fast ionization wave (FIW). In the years following its discovery, a substantial effort was made to document the properties of the FIW over a wide range of conditions. In these studies, the wave velocity, current, and attenuation were the most frequently measured quantities. Much of this work is summarized in a review by Vasilyak [38]. Also reviewed are Slavin and Sopin's work which was the first to attempt a computation of the electron energy distribution function EEDF in FIWS [39].

The experimental measurements and computational work reported by Vasilyak were expanded on by a series of studies conducted at the Moscow Institute of Physics. These are reviewed by Starikovskaia et al. [40] and included measurements of the electron density, electric field, and energy coupling for FIWS in air, nitrogen, and hydrogen. The computational work by Starikovskaia and Starikovskii [41] still represents the most detailed study of the EEDF in nitrogen FIWS.

However, Starikovskaia et al. noted that the usefulness of FIWS were limited, in part, by their repetition rates. The power supplies for FIWS were capacitor

banks, charged in parallel, and discharged in series (also referred to Marx banks). Unfortunately, the spark gaps used to trigger these capacitor banks would not operate above a few hundred Hz. This changed in the late 1990's with the development and commercialization of fast, solid-state switches. Specifically, with the fast ionization dynistor it was possible to achieve repetition rates of 100 kHz.

This led to a new class of repetitively-pulsed discharges, or the RPND. These discharges operated at sufficiently high rates such that charged particles would persist between pulses, significantly increasing the plasma duty cycle. The improved qualities of the RPND over the FIW inspired a number of novel, application-driven studies. This included:

- Plasma-assisted combustion [42–44]
- Magnetohydrodynamic energy bypass engines [44–46]
- Plasma actuators [47, 48]
- High-pressure xenon lamps [49]
- Plasma medicine [50, 51]
- Water treatment [52]

Though not specific to the RPND, Becker et al. [53] provide an extensive discussion of the potential uses for non-equilibrium air plasmas.

As a result, contemporary researchers have produced a wealth of literature on the operation of RPNDS. More recently, there have been detailed measurements of

the gas temperatures [42, 54–60], chemical composition [56–58], electric fields [61–63], and energy coupling [42, 64]. Notably, these studies have been generally restricted to molecular gases; air, nitrogen, and occasionally, hydrogen.

The first such study was the work of Laroussi and Lu who examined a RPND excited in a stream of helium flowing from a tube into air [65, 66]. The resulting plasma had the appearance of a jet, emitted from the open end of the tube. Using fast photography they observed that the jet was actually a series of plasma “bullets” formed with each pulse. Measurements of the bullet velocities showed that their speed greatly exceed what would be expected purely from electrons drifting under the applied electric field. They described the bullet as a classic cathode-directed streamer propagated by photoionization.

The plasma bullets of Laroussi and Lu spawned a great deal of interest in RPND helium plasma jets<sup>3</sup> For example, Walsh et al. studied the atomic oxygen production for helium-oxygen mixtures with the use of emission spectroscopy and a global plasma chemistry model [69]. Urabe et al. employed a variety of laser diagnostics to measure the radial density profiles of helium metastable atoms and molecular nitrogen ions in a similar jet. This work was supported by a number of two-dimensional plasma simulations such as those by Naidis [70] and Breden, Miki, and Raja [71].

Simultaneously, there has been a decline in the study of FIWs, and relatively little on large-volume RPNDs. One of the most recent FIW studies was produced by

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<sup>3</sup>A distinction should be made between plasma jets, excited by sinusoidal power supplies, similar to the well-known dielectric-barrier discharge [67], and those produced by nanosecond pulses. Differences between the two were reported by Walsh, Shi, and Kong [68].

Takashima et al. [72]. In it, the authors reported on FIWs in helium and nitrogen which were studied using capacitive probes and voltage-current characteristics. The results were compared to extensive two-dimensional fluid simulations and an analytic, one-dimensional drift model. In most cases, the measurements and simulations showed good agreement.

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