Chapter 1

Introduction

1.1 Overview

Historically, the study of atmospheric-pressure plasmas (APP's) is indistinguishable from the study of plasmas as a whole. However, the detail of the measurements and calculations associated with APP's has been limited by their complexity. From a computational perspective, the high pressure and number of potential reactions present a difficult challenge. Likewise, the high pressure can significantly complicate the data analysis for a number of plasma diagnostics. Aside from the high pressures, the large electric fields, short time scales, and general randomness of APP's make even the most basic observations a feat.

In the last several decades, some of this has begun to change. High-powered computing has allowed simulations with remarkable detail.

Similarly, advances in technology has enabled plasma diagnostics in regimes that were experimentally inaccessible. As a result, the body of knowledge regarding APP's has greatly increased. Sometimes, the motivation for this work is scientific curiosity. More often, the study of APP's has been driven by a broad range of applications.

Among the first plasma applications were provided by APP's: ozone generation and lighting. Aside from these items, plasma welding, polymer treatment, combustion, and plasma televisions have become widely accepted. However, a large number of new applications may soon be added to this list, including: treatment of tissue wounds, altering airflow over airfoils, and destruction of industrial pollutants.

Unsurprisingly, each case has demands a different kind of plasma. The original arc discharges were created between two graphite rods connected to immense battery banks. In contrast, a modern research reactor studying plasma-assisted combustion might use a fast-switching semiconductor circuit. Over the years, several types of APPS have been developed for a variety of situations: dielectric-barrier, corona, thermal arc, RF, microwave, pulsed, and more.

Within this group[1], the repetitively-pulsed nanosecond discharge (RPND) has created considerable interest. Generally speaking, a RPND is a plasma generated by a repetitive electrical pulse applied between two electrodes. The pulse voltage if often > 1 kV, lasts anywhere from

[1]The interested reader is referred to Starikovskaia's review [1] which provides a general overview of APPS in the context of plasma-assisted combustion

< 1–100 ns, and is repeated over a thousand times each second. The result is a wave of ionization (and light) which crosses from the powered electrode to the grounded one.

A RPND can fill volumes of several liters with a relatively uniform plasma. Though they can cause significant excitation of the background gas, they generally produce very little heating (in some cases no more than a few kelvin above room temperature). In addition, the excitation can be changed with adjustments to the magnitude or duration of the electrical pulse. Each of these characteristics are highly desirable in one or more of potential applications for APP's.

Given all of these promising properties, RPND's have been the subject of substantial study by several research groups. However, much of this work has focused on the physics of RPND's in air. Unfortunately, air's large number of constituent elements can lead to notable complexity. In turn, this can obscure some of the more fundamental questions relation to RPND's: how do they form, how is the energy distributed between excited particles, what kind of spatial variation can be expected?

This paper details a study of each of these questions in a helium RPND. Specifically, the densities of one particular excited atom are measured for a variety of pressures and locations. This is complemented by measurements of the light emissions for the same set of parameters. A simple model of a RPND is used to predict several characteristics of the plasma based on the excited state densities: electron density, electric field, and light emission. The measured light emissions are interpreted to show how the energy is distributed in the gas, and how it changes over time. Finally, they are compared with the estimated light emissions to check the validity of several common assumptions.

The remainder of this chapter is comprised of a review of the associated literature, as well as a discussion of basic discharge theory. Chapter **??** covers the experimental setup as well as some general observations of the RPND. Next, the measurement of the excited state densities is presented, followed by the chapter on the light emission measurements. Chapter **??** explores the global model used to interpret the excited state densities, as well as some supporting particle-in-cell simulations. Finally, the paper concludes with a discussion of how the models and measurements impact the present understanding of RPND's.

1.2 Literature Review

History of Atmospheric-Pressure Discharges

Like most physical phenomena, plasmas are typically only described under ideal circumstances. This means that neutral collisions, and subsequently, atmospheric plasmas, are often ignored. Neutral col-

Figure 1.1: This is a test.

lisions compete with the electromagnetic effects that distinguish a plasma from a gas. Therefore, they're an undesirable complication in most academic discussions. Of course, as was mentioned, the history of plasmas is very much defined by the study of APP's. Aside from stars, the first plasmas seen by man were almost certainly lightning and static sparks (both of which, fall under the category of APP). Furthermore, the first artificial plasmas were arcs in atmosphere, and can be attributed to Vasilii Petrov and Humphry Davy [2].

The research of Petrov and Davy is the first work on a type of plasma that is now referred to as a thermal arc. Such plasmas are common in industrial lighting systems, and for a time were even used as the primary nigh-time illumination of Detroit. Perhaps more common, is the use of thermal arcs for the welding or cutting of metals. This potential was recognized early on. Volta's recent invention of the voltaic pile provided the first source of constant and sufficient electrical energy. Using a series of voltaic cells, Petrov was able to draw the first electrical arc between two sticks of carbon. Its blinding light was recorded in a number of historical prints, such as figure 1.2. Aside from this light, the arcs were characterized by their significant ionization, and high degree of thermal equilibrium. Gas temperatures could reach thousands of kelvin.

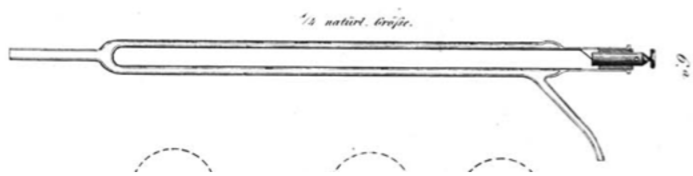

Figure 1.2: Werner von Siemens' silent discharge. Per Rackham requirements, the caption text should be singled-spaced, regardless of the text body. I'm giving this a test right here. Yep. It should stay single-spaced. Pretty please?

Both the primary advantage and disadvantage of thermal arcs are their high operating temperatures. These high operating temperatures correlate with the ability of the plasma to convert electrical energy to thermal energy; of great values in welding or for continuum light generation. In these plasmas, some of the energy is also converted to excited atoms or molecules. In some cases, these species may be desirable for processing a material. However, contact with these high temperatures will often lead to destruction of the substrate.

In contrast, later work by Werner von Siemens [3], led to the discovery of the so-called "silent discharge," seen in figure 1.2. In recent years, the terminology has changed and this type of discharge is now referred to as a dielectric-barrier discharge, or DBD. The DBD was significantly different from the thermal arc. Visually, it was much dimmer, and appeared to be composed of many thousands of individual filaments. Additionally, the DBD did not significantly heat the air, unlike the thermal arc. Finally, the DBD was used in the first commercial plasma application: ozone generation and water purification. Notably, both the thermal arc and silent discharge predated the 'official' discover of plasma by Sir William Crookes in 1872.

The DBD could be used process materials. Indeed, it has become common to use this type of discharge to treat polymer films, as well as clothes, and medical equipment (sterilization). However, like the thermal arc, there are some limitations. DBDs are not particularly uniform as a result of the large numbers of filaments. Thus, uniform processing will only occur as a result of long processing times–when the relatively random position of these filaments is averaged out–or if the desirable species has a long enough life to diffuse away from where it was created. Another limitation to DBDs is that the filaments characteristics are not a strong function of the applied voltage. This leads to limited flexibility in control of the plasma characteristics.

Nanosecond-Pulsed Plasmas

For a substantial period of time, these two discharges represented the range of atmospheric-pressure plasmas (APP). The thermal arc, though useful, could not be used on delicate substrates. It had the additional problem of having relatively little control over its chemical kinetics. Meanwhile, the DBD was relegated to ozone production and polymer processing (relatively low-value applications). Though the DBD had attractive thermal properties, little else was known about how it op-

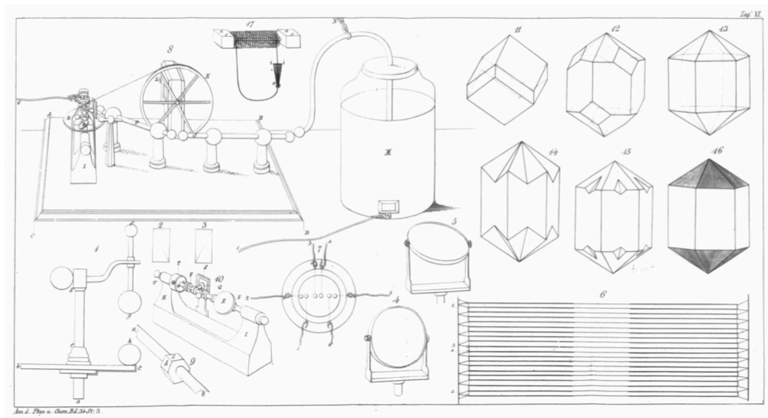

Figure 1.3: Sketches of the discharge apparatus and measurement system used by Wheatstone.

erated, and how to control its properties. As recently as 2007, the National Academies noted 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, likened to aerodynamics." [4]

As with the discovery of the thermal arc and the dielectric barrier discharge, the first pulsed discharges were studied prior to the official discovery of plasma. In 1835, Charles Wheatstone attempted to measure what he called the velocity of electricity in a spark gap, powered by a Leyden jar [5]. In hindsight, the question was poorly phrased; Maxwell's equations had not yet been formalized, and electrons weren't recognized as the carriers of electrical current. Nevertheless, though poorly controlled, the phenomena was essentially the same as modern pulsed nanosecond discharges.

Conceptually, the work of Wheatstone was promising, however the measurements were quite inaccurate. One particularly important outcome of Wheatstone's work was the subsequent observation by von Zahn [6] that the particles emitting the light were not travelling at anywhere near the velocity of the wave of light. J.J. Thomson later repeated the experiment with a 15 m tube, in order to obtain an improved estimate of the wave velocity and its direction [7]. He estimated the velocity of the wave to be approximately one half the speed of light, traveling from the anode to the cathode.

These high velocities caused an moderate amount confusion, particularly because there appeared to be no associated motion of the emitting particles. Subsequent examinations be Beams [8] confirmed the velocity measurements, and more importantly demonstrated that, regardless of the polarity of the applied potential, the luminous wave always travelled from the high potential to the low potential electrode. He additionally noted that the large pulse of current associated with such waves was not detected until after the wave had crossed the length of the tube. This observation led to the astute observation by Beams that the apparent motion of the luminous front was more likely a moving region of ionization.

As observed by Loeb [9] in his unifying description of pulsed nanosecond discharges, aside from the group of people studying the propagation of light in rarefied gas discharges, were a different set of scientists working on the origin of lightning. The endeavor was ambitious, for many of the same reasons that atmospheric discharges have always been difficult to study. Basically, no one was quite sure where to point their cameras, and when to open the shutters. Though out of the scope of this paper, the information gleaned from these studies was both relevant and useful in the development of the theory underlying ionization waves. Interested readers are referred to the review by Gurevich and Zybin [10] for an overview of the models employed in describing natural lightning.

Though studies continued intermittently after a burst of interest in the 1930s, it wasn't until the mid-1960's and 1970s when any significant attention came to the pulsed-nanosecond discharge. Beginning with Loeb's description of the phenomena as "ionizing waves of potential gradient," several more articles appeared exploring the nature of these pulses. Mesyats, Byckhov, and Kremnev [11] developed a theory explaining the breakdown process which is among the first to specifically reference runaway or nonlocal electrons. This theory was continued with a kinetic treatment and several simulations by Kunhardt [12], and Kunhardt and Tzeng [13].

Research Plan

Propose research to fill this gap Specific and cited history of PNDS and related measurements.

1.3 Basic Theory

Basic theory of gaseous breakdown.

Bibliography

[1] S M Starikovskaia. Plasma assisted ignition and combustion. *Journal of Physics D: Applied Physics*, 39(16):R265–R299, August 2006.

[2] André Anders. Tracking down the origin of arc plasma science-II. early continuous discharges. *IEEE Transactions on Plasma Science*, 31(5):1060–1069, October 2003.

[3] W. Siemens. Ueber die elektrostatische Induction und die Verzögerung des Stroms in Flaschendrähten. *Annalen der Physik und Chemie*, 178(9):66–122, 1857.

[4] Plasma 2010 Committee, Plasma Science Committee, and National Research Council. *Plasma Science: Advancing Knowledge in the National Interest*. Number 2007. The National Academies Press, Washington, D.C., 2007.

[5] C. Wheatstone. Versuche, die Geschwindigkeit der Elektricität und die Dauer des elektrischen Lichts zu messen. *Annalen der*

Physik und Chemie, 110(3):464–480, 1835.

[6] W. v. Zahn. Spectralröhren mit longitudinaler Durchsicht. *Annalen der Physik und Chemie*, 244(12):675–675, 1879.

[7] J J Thomson. *Notes on Recent Researches in Electricity and Magnetism*. Clarendon Press, Oxford, UK, 1893.

[8] J. Beams. The Propagation of Luminosity in Discharge Tubes. *Physical Review*, 36(5):997–1001, September 1930.

[9] L B Loeb. Ionizing Waves of Potential Gradient: Luminous pulses in electrical breakdown, with velocities a third that of light, have a common basis. *Science (New York, N.Y.)*, 148(3676):1417–26, June 1965.

[10] Aleksandr V Gurevich and Kirill P Zybin. Runaway breakdown and electric discharges in thunderstorms. *Physics-Uspekhi*, 44(11):1119–1140, November 2001.

[11] Gennadii A Mesyats, Yu I Bychkov, and V V Kremnev. Pulsed nanosecond electric discharges in gases. *Soviet Physics Uspekhi*, 15(3):282–297, March 1972.

[12] E. Kunhardt and W. Byszewski. Development of overvoltage breakdown at high gas pressure. *Physical Review A*, 21(6):2069–2077, June 1980.

[13] Ee Kunhardt and Y Tzeng. Development of an electron avalanche and its transition into streamers. *Physical Review A*, 38(3):1410–1421, August 1988.