Population Kinetics of a Repetitively-Pulsed Nanosecond Discharge

by

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I would like to dedicate this dissertation to someone else.

ACKNOWLEDGMENTS

Who is this?

Preface

This is a dissertation about something; I really hope it's good.

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DBD dielectric-barrier discharge

APP atmospheric-pressure plasma

CHAPTER 1

Introduction

1.1 Overview

Research on atmospheric plasmas has been going on for as long as research on plasmas themselves. However, it was only recently that the necessary diagnostic techniques and computational power existed to properly examine them. In the last few decades there has been a renaissance in work dedicated to atmospheric plasmas, particularly atmospheric pressure plasmas that are out of thermal equilibrium. This is because they are able to treat delicate substrates with none of the thermal damage associated with arcs.

Though several approaches exist to generating atmospheric pressure plasmas (dbd, microwave, rf), particularly interesting are those using nanosecond pulses. The duration of such pulses, as well as their amplitude can be changed to target particular atomic or molecular reactions. This flexibility is highly desirable in the world of plasma processing where selectivity, control, etc. are of utmost importance. However, we are still learning how to measure these particular plasmas. As recently as 1994 [1] the only diagnostic with any meaningful accuracy was propagation velocity.

Several research groups have focused on the development of pulsed nanosecond discharges in air. However, the complex chemistry associated with air plasmas obscures some of the more fundamental questions: how is the pulsed nanosecond plasma formed, how are the excited states of the system populated, what significance is there to reactions after the fast ionization wave, the spatial variation of system parameters, what kind of electron energy distribution can be expected? This study will emphasize spectroscopic measurements of a helium pulsed nanosecond discharge.

Such a system retains physical relevance given that helium is a common stabilizing additive to atmospheric pressure plasmas. At the same time, the more simple atomic structure lends itself to a more detailed examination using global models, kinetic simulations, and active spectroscopy. This work will focus its efforts on the description of the spatial variation of the pulsed-nanosecond discharge, an approximation of its electron energy distribution function, and a description of how the neutral atoms are excited.

The remainder of this chapter includes a review of the associated literature, as well as a discussion of basic discharge theory (diagnostic-specific theory will accompany the relevant chapters). Chapter 2 provides the necessary details of the experimental setup as well as some preliminary observations. Next, in chapter 3, absolute measurements of the helium triplet metastable densities are presented. Chapter 4 covers more general emissions measurements. In order to provide a more clear understanding of the meaning of these measurements, we explore them using a global model and a particle kinetics model. Finally, we conclude in chapter 6 with a discussion of how the collection of models and measurements influences our understanding of the pulsed nanosecond discharge.

1.2 Literature Review

1.2.1 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 collisions tend to obscure the electromagnetic effects that distinguish a plasma from a gas. However, the history of observation and study of plasmas is indelibly linked to atmospheric plasmas. Lightning and static sparks are the most prevalent plasmas on earth. Indeed, the among the first artificial plasmas was a constant atmospheric arc generated in 1802, the work of a Russian scientist named Vasilii Petrov [2].

Indeed, at present, there's a relative abundance of different atmospheric pressure discharges: dielectric-barrier, RF, microwave, glow, pulsed, and more. Each has unique range of parameters in which it operates, and a survey of all possible atmospheric discharges would be excessive. Instead, we shall focus on the two most common discharges which, together, illustrate the distinct advantages of the nanosecond-pulsed discharge.

The work of Petrov was the forerunner to what is now referred to as the study of thermal arcs. 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 1.2.1. Aside from this light, the arcs were characterized by their significant ionization, and high degree of

Figure 1.1: This is a test.

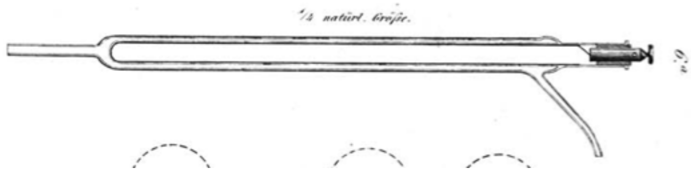

Figure 1.2: Werner von Siemens' silent discharge. Per Rackham requirements, the caption text should be singled-spaced, regardless of the text body. I'm giving this a test right here. Yep. It should stay single-spaced. Pretty please?

thermal equilibrium. Gas temperatures could reach thousands of kelvin.

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 1.2.1. 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.

1.2.2 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 operated, 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." [**?**]

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 [**?**]. In hindsight, the question was poorly phrased; Maxwell's equations had not yet been formalized, and

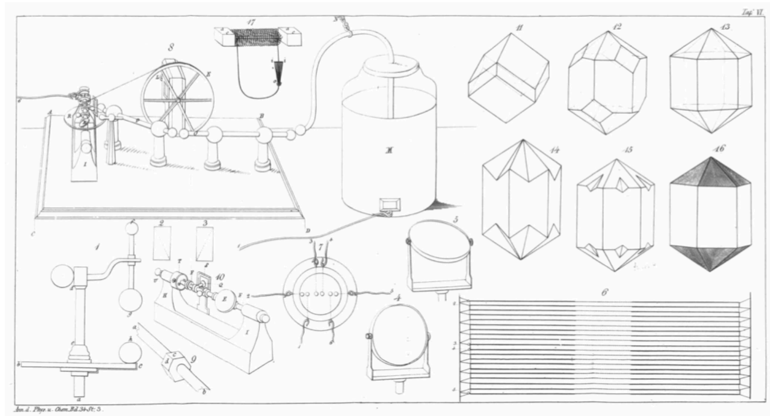

Figure 1.3: Sketches of the discharge apparatus and measurement system used by Wheatstone.

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 [**?**] 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 [4]. 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 [**?**] 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 [5] 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 [**?**] 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 [6] 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 [**?**], and Kunhardt and Tzeng]citeKunhardt1988.

1.2.3 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.

CHAPTER 2

Experiment

CHAPTER 3

Metastable Measurements

CHAPTER 4

Emission Measurements

CHAPTER 5

Modeling

CHAPTER 6

Conclusions

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