Chapter 1

Introduction

1.1 Overview

Answer this question: how many references do you want here? How many are you going to delay until the lit review?

Short paragraph, non-rigorous definition of plasmas. Be sure to clarify distinction from electrical discharge, or if you consider them the same.

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.

Ambiguous start to paragraph, specify and cite problems, then identify how they have been overcome.

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. Meanwhile, 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 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 is often in excess of one kilovolt, lasts anywhere from $< 1 - 100$ 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 below a detection limit of $\Delta \pm 15$ K). 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 dis-

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

tributed between excited particles, and 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.

Paragraph on reason for choosing helium, specify choice of measurements and topic of dissertation

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

Though RPND's are very much a product of twentieth century research, they are fundamentally similar to a number of other pulsed discharges such as electrical sparks and lightning. Though Loeb united these disparate fields under the title of "ionizing waves of potential gradient" in 1964 [2] (we use the more familiar term, fast ionization waves), the underlying subjects had been under study since the Greeks who generated sparks by rubbing together amber and fur.

Despite these early observations, it was Leibniz in 1671 who first came to the conclusion that sparks were an electrical phenomena [3]. Subsequently, Franklin's famous kite experiment led him to a similar conclusion on the nature of lightning. Franklin was also involved in explaining the principles of Leyden jars, develop by Musschenbroek. The Leyden jar was the first reliable way to store electrical energy and proved a boon to later research.

In 1835, Wheatstone made the first attempt to measure the speed of electricity through a gas [4]. In his work, Wheatstone used a Leyden jar connected to two metal spheres, separated by a small gap. Once the charge in the jar reached a critical level, a spark would form in the

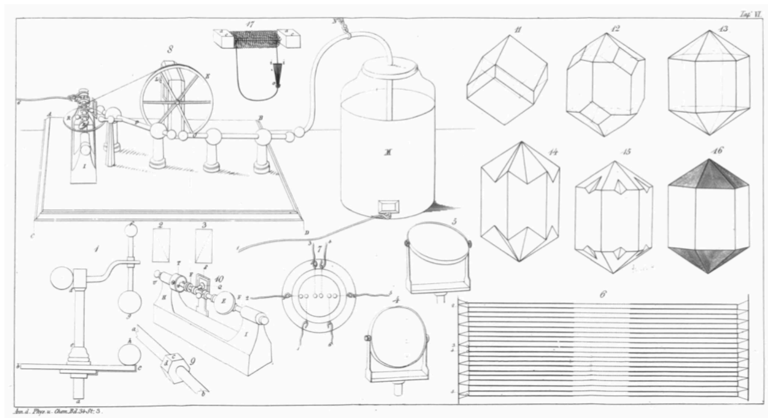

Figure 1.1: The experimental sketches of Wheatstone showing a traditional spark gap connected to a Leyden jar and electrostatic generator.

gap. Figure 1.1 shows the experimental sketch provided by Wheatstone. Though the measurement is notable for its early date, it was later revisted with much more accuracy by Thomson [5]. Perhaps the most important outcome of Wheatstone's study was the observation by Zahn [6] that the speed of the light was *not* accompanied by a similar motion of the emitting particles.

Thomson's work concerned both the speed and direction of light in a pulsed discharge. Unlike Wheatstone's study, Thomson used an elongated tube, 15 m in length, and 5 mm in diameter, upon which he drew a vacuum. The original sketch of Thomson's discharge apparatus can be seen in figure 1.2. Through a clever arrangement of mirrors, Thomson determined that the electricity had a speed approaching 1×10^{10} cm/s, and travelled from the anode to the cathode.

It was later, in 1930, that Beams would determine that the wave

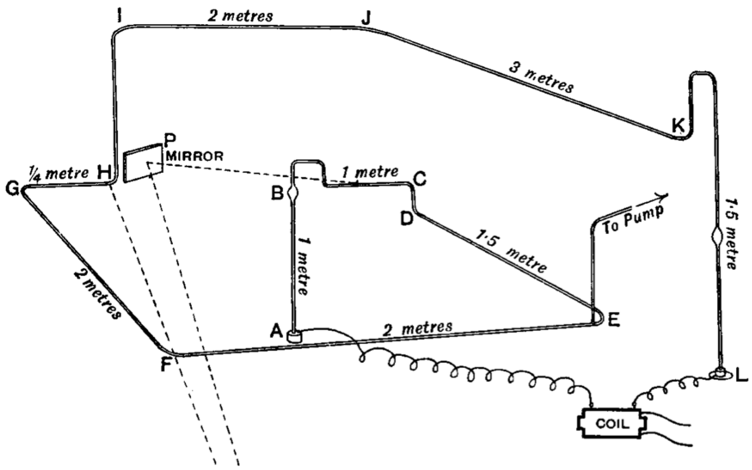

Figure 1.2: A sketch of J.J. Thomson's early experiments on fast ionization waves in long vacuum tubes.

always initiated at the high voltage electrode, regardless of polarity [7]. In addition, Beams measured the current at the low potential electrode. He detected a current pulse which did not appear until after the light had completely crossed the gap. He came to the conclusion that the luminous front was likely the result of a moving region of ionization.

Around the same time, there was a distinct set of researchers ho were studying similar phenomena in lightning. In most cases, these studies concentrated on time-resolved photography, pioneered by Boys[2][8], and refined by Schonland [9]. This technique was later adopted by Allibone and Meek [10] to observe the evolution of a laboratory-generated

[2]In the same article, Boys anticipated a number of other atmospheric physics studies by proposing that rockets be fired at thunderclouds. Unfortunately, he lived in a village of thatched houses and could not conduct the experiment for fear of fire.

spark.

Perhaps address the theory of rpnds separately?

By 1935, fast ionization waves had been under study for nearly 50 years. However, there was still no adequate explanation for the speed of the discharge. Similarly, Beams' observation that the wave always travelled from the high voltage to the low voltage electrode (regardless of polarity) was inconsistent with the Townsend model used to explain most plasmas. Based on observations made with fast pulses, Flegler and Raether developed a new theory of breakdown for sparks in air [11] which was capable of, at least partly, explaining the fast ionization wave phenomena. Independently, Loeb and Meek developed a similar theory in 1940 [12].

The work of Fleger and Raether as well as Loeb and Meek, was intended to explain the breakdown processes for an undetermined range of overvoltages (in excess of the breakdown voltage). However, as early as 1951, Fisher and Bederson [13], demonstrated that the Townsend mechanism was still plausible at low overvoltages.

In contrast, 1961 saw Fowler [**?**] seeking a hydrodynamic explanation for the waves. Contrary to the stochastic and particle-based description of the streamer breakdown, Fowler considered the luminous fronts to be nonlinear electron acoustic waves. Though the explanation provided a fair agreement with his observations, there were several issues with the analysis, namely a simplified consideration of the geometry and associated field strengths.

By 1965, Loeb himself, admits that photoionization was not insufficient on its own to produce the observed phenomena. In his review for Nature, Loeb identified several phenomena that exhibited similar characteristics. The return stroke in lighting, high overvoltage breakdown in rarefied gases, and sparks in atmosphere. Loeb was able to provide a qualitative description of the physics involved, but ultimately deferred on any quanitative description.

The insufficiency of photionization was later reinforced by the observation of Mesyats [14] that the speed of the discharge processes was often faster than the lifetimes of excited states. Again, this precluded photoionization from providing a significant amount of preionization for the propagation of an rpnd. Mesyats instead suggested that the large fields generated an electron avalanche that grew much more rapidly than the typical Townsend discharge. This was followed by an avalanche chain which further propagated the plasma.

Later, Kunhardt [15] extended on Mesyats' analysis and provided a more theoretical underpinning for it. Taking his inspiration from the group theory used in neutron diffusion, Kunhardt explored the development of a fiw from the perspective of “trapped” and “runaway” electrons. Previous work by Babich and Stankevich [**?**] inspired this by suggesting the existence of continually-accelerated electrons at high overvoltages.

As an aside, the topic of electron beams in rare gases became of substantial interest to researchers in the mid 1970s. Because rare gases lacked low-lying excited states which might detrimentally absorb energy, they were favor for laser where a population inversion could be achieved with ease. As a result, a great deal of work went into detailing the propagation of an electron beam in rare gases which is physically similar to the development of a fast ionization wave.

It was around this time that the topic of fiw became of substantial interest to Russian research groups. Though much of the early work is shrouded in the mists of language differences, it is believed that Vasilyak [16] provides a fair review of the material.

Come 1998, the fiw was the subject of renewed interest by a group of researchers at the Moscow Institute of Physics and Technology [**?**]. They employed several different diagnostic techniques (photomultiplier tubes and capacitive probes) in an exceptionally detailed study of fiws in a shock tube. Initially, the work focused on the decay of excited states in nitrogen. As

Remember to include the laser dudes! Xenon lamps!

1.3 Basic Theory

Plasmas

A volume containing some number of charged particles can be considered a plasma if it meets three conditions. The first requires that the motion of charged particles is primarily determined by the electric and magnetic fields of the volume rather than through collisions with neutral particles. This is classically expressed by the inequality

$$\sqrt{n_{\mathrm{e}} e^2/(\epsilon_0 m_{\mathrm{e}})} < \nu, \tag{1.1}$$

where n_{e} is the electron density, e is the fundamental charge, ϵ_0 is the permittivity of free space, m_{e} is the mass of an electron, and ν is the electron-neutral collision frequency. The left-hand side term is called the electron plasma frequency, it the characteristic frequency at which a plasma oscillates in response to a perturbation.

requirements for a plasma

Be clear about relation to Lieberman's writing!

For a sufficiently large number of particles, the behavior of the each species of the plasma can be described by a continuous probability distribution function. This function, $f_\alpha(\vec{r}, \vec{v}, t)$, describes the probability of finding a particle of species α, at position $\vec{r}$, The distribution function for a particle can be determined by the Vlasov-Fokker-Planck **vfp!**

equation,

$$\frac{\partial f_\alpha}{\partial t} + \vec{v_\alpha} \cdot \nabla f_\alpha + q_\alpha \left(\vec{E} + \vec{v_\alpha} \times \vec{B} \right) \cdot \nabla_{\mathrm{v}} f_\alpha = \left(\frac{\partial f_\alpha}{\partial t} \right)_{\mathrm{coll}} . \tag{1.2}$$

Here, $\vec{E}$ is the electric field, $\vec{B}$ is the magnetic field, and $\partial f_\alpha/(\partial t)_{\mathrm{coll}}$ is a term representing all collisions. The **vfp!** equation is coupled to Maxwell's equations in order to obtain a self-consistent description of the particle distribution and the resulting fields. In essence, this is the Boltzmann equation from statistical mechanics, however it now includes several changes. Vlasov replaced the original force term with the Lorentz equation, and Fokker and Planck introduced the collision operator on the right-hand side. This is coupled with Maxwell's equations for a solution of the electric and magnetic fields in the plasma.

In the absence of external fields and with only elastic collisions, the equation admits the famous Maxwell-Boltzmann equilibrium distribution,

n for degrees of freedom? Change to something unambiguous?

$$f_\alpha(v) = n \left(\frac{m_\alpha}{2\pi k_{\mathrm{B}} T} \right)^{3/2} \exp \left(-\frac{m_\alpha v_\alpha^2}{2 k_{\mathrm{B}} T} \right) , \tag{1.3}$$

where n is the number of degrees of freedom, k_{B} is Boltzmann's constant, and T is the temperature. A species of particles which possesses a Boltzmann distribution is said to be in equilibrium. Likewise, two species with the same distribution are in equilibrium.

Should the Boltzmann-Maxwell equilibrium be pushed back to discussion of rate equations and averaging?

Aside from this, the **vfp!** equation is notoriously difficult to solve. As a result, most plasma models use various moments of equation 1.2 where the velocity dependence has been integrated out. These moments are the basis for the two-fluid equations, the MHD formulation, and global models. We will show the first three moments following the notation of Lieberman and Lichtenberg [17]. For example, the first moment is the continuity equation,

$$\frac{\partial n_\alpha}{\partial t} + \nabla \cdot (n_\alpha \vec{u_\alpha}) = G_\alpha - L_\alpha, \tag{1.4}$$

where $\vec{u_\alpha}$ is the mean velocity of species α, G_α is its rate of gain, and L_α is the rate of loss. This equation can be interpreted as the rate of change in particle density for a particular volume of space.

Though the continuity equation is much simpler than the original **vfp!** equation, it cannot be solved alone. The mean velocity, $\vec{u}$, is undefined. Typically, this leads to the second moment,

Come up with tensor notation, and fix collision operator

$$mn_\alpha \left[\frac{\partial \vec{u_\alpha}}{\partial t} (\vec{u_\alpha} \cdot \nabla) \right] = q_\alpha n_\alpha (\vec{E} + \vec{u_\alpha} \times \vec{B}) - \nabla \cdot \vec{\Pi_\alpha} + \vec{f}_{\alpha,\text{coll}} \tag{1.5}$$

where $\vec{\Pi_\alpha}$ is the pressure tensor, and $\vec{f}_{\alpha,c}$ is the rate of momentum transfer into species α. Again, any solution is stymied by the presence of a new a term, in this case, $\vec{\Pi}_\alpha$. At this point, an equation of state can be used to close the set of equations, in this case relating the pressure to the density. However, later work will benefit from one more moment.

Following the conservation of momentum, the energy conservation equation can be derived from the **vfp!** equation,

$$\frac{\partial}{\partial t}\left(\frac{3}{2}p_\alpha\right) + \nabla \cdot \frac{3}{2}(p_\alpha \vec{u}_\alpha) + p_\alpha \nabla \cdot \vec{u}_\alpha + \nabla \cdot \vec{q}_\alpha = \frac{\partial}{\partial t}\left(\frac{3}{2}p_\alpha\right)\Big|_{\text{coll}} \tag{1.6}$$

where p_α is the species pressure, q_α is the heat flow vector, and the right-hand side is the time rate of change in energy as a result of collisions. In our case, we only consider the flux into the volume (from the electric field) and the distribution of this field via rate constants. This is the basis for the global model.

Atomic Spectroscopy

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