Abstract of “Studies of Negative and Positive Charge Carriers in Superfluid Helium-4” by Stephen B. Sirisky, Ph.D., Brown University, May 2019

Charge carriers have long been introduced into superfluid helium-4, and their mobilities measured. Electrons repel the atoms of the liquid in their vicinity, forming a small cavity, or “bubble”. Of particular interest are the “exotic ions”, which have mobilities larger than that of the electron bubbles. By measuring the mobilities of different charge carriers we can deduce information about the microscopic structures they form in the liquid. Four different experiments were performed.

In the first experiment light of wavelength 1064 nm was used to generate optical breakdown of the helium liquid. The threshold intensity was measured over the temperature range from 1.1 to 2.8 K. Experiments were performed to study how the breakdown from one pulse modifies the probability that a subsequent pulse will result in breakdown.

In the second experiment the “fast ion”, the fastest of the exotic ions, was generated with an Americium-241 source. The experimental cell was pressurized up to 5.5 bar and the mobilities of the fast ion and normal electron bubble were measured. From these measurements we study how the radius of the fast ion changes with increasing pressure.

In the third experiment the “#2 background”, a signal from charge carriers which are continuous in mobility, was studied. This background is interpreted as electrons from the discharge which have energy above that of the barrier for entry into liquid helium, but are only partially transmitted across the vapor-liquid interface. The transmitted portion of the electron wave function is trapped, forming a bubble which contains only a fraction of the wave function, and is thus smaller than the normal electron bubble. The experiment shows that the interaction of the electron with the liquid helium does not result in a measurement that quickly determines that an electron is in the bubble or is not in the bubble.

Finally, in the fourth experiment observations of new positive charge carriers with mobilities close to that of the normal He⁺ snowball are detailed. The temperature and field dependence of their mobilities and amplitudes in the region of 1 K are measured.
STUDIES OF NEGATIVE AND POSITIVE CHARGE
CARRIERS IN SUPERFLUID HELIUM-4

by

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For my loving mother Lisa, father Scott, 
and sister Amanda 
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To my favorite cousins, Adam, Luke, Sofia, and Brooke
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Chapter 1

Introduction

In 1908 Kamerlingh-Onnes was the first person to liquefy helium. This result led him to discover superconductivity in a related experiment soon after (1911). These major steps paved the way for modern low temperature research. Helium in the liquid phase exhibits a two phase structure. There is the normal phase (I), which as the name implies behaves like a typical fluid, and the superfluid phase (II), which has no viscosity and very high conductivity. The transition between the two phases at saturated vapor pressure (SVP) occurs at a temperature $T_\lambda = 2.17 \, K$. This is illustrated in the helium phase diagram in the left panel of Fig. 1.1. The transition temperature is referred to as $T_\lambda$ due to the $\lambda$ like shape of the helium heat capacity near this point. The dispersion relation for the superfluid, shown in the right panel of Figure 1.1 can be split up into two different regions. At low wave number the dispersion relation is dominated by a linear relation, $\epsilon(k) = c$ $\hbar$ $k$, where $c$ is the sound velocity, and so these excitations are referred to as phonons. Around the minima the energy of excitations depends quadratically on the wave number, $\epsilon(k) = \Delta + \frac{\hbar^2}{2m} (k - k_o)^2$, where $m$ is the effective mass of the excitation. These are referred to as rotons, and the height of the minima $\Delta$ the roton energy gap.

At low temperature helium forms a Bose Einstein condensate (BEC). Since the
helium atoms are in their lowest energy state, any loss of energy needs to occur via production of an excitation. Conservation of energy and momentum demands that if a collision of an object with a medium is to produce such an elementary excitation, it is only possible when the velocity of the object is at least \( v_c = \left( \frac{E}{p} \right)_{\text{min}} \), the Landau critical velocity. In the linear phonon region of the dispersion relation, \( E/p = c \). In the roton regime, however, \( v_c \approx \frac{\Delta}{\hbar k} \). \( v_c \) is approximately 60 m/s, which happens to be less than \( c \). Thus, at velocities below \( v_c \), objects are unable to lose energy to excitations and therefore experience no viscosity. These properties make liquid helium ideal for studying low temperature quantum phenomena, and have motivated decades of experiments.

At finite temperature, collisions with these excitations cause ions to experience a drag force. As a result, under the influence of low electric fields \( E \) the ions accelerate until they reach a maximum drift velocity, \( v_d = \mu E \), where \( \mu \) is the ion mobility. Low field strengths are considered to be those for which the velocity is below that required for the onset of critical phenomena, such as vortex nucleation.

A series of experiments performed in the 1950s by Meyer and Reif [10,11,12] and Schwarz [13] measured the mobilities of the positive and negative charge carriers over
a large range of conditions, including temperature, electric field, and pressure. At constant pressure the mobilities of both species were found to vary near $T = 1$ K as $\mu \propto e^{\Delta/kT}$, consistent with a velocity determined mainly by the frequency of collisions of rotons with the ions. They found that the magnitudes of the positive and negative ion mobilities differ by significant amounts, at least 50% from 0.8K to 1.2K. A more detailed theory for the ion mobility that was found to agree well with experiment was later formulated by Barrera and Baym [14].

Around the same time, two ideas for the ionic structures took shape. In 1959 Atkins [15] proposed that a positive ion, by polarizing the nearby neutral helium atoms, could produce pressures well in excess of the freezing pressure via electrostriction. The ion, surrounded by solid layers of atoms from the bulk, was thus coined the ‘snowball’. His initial calculation estimated a snowball radius of approximately 7 Å.

Ferrell [16], and then Careri [17, 18], proposed that a lone electron might find it energetically favorable to “push” away the surrounding helium atoms and localize within a small region. To enter the bulk, electrons have to overcome a 1 eV potential barrier [19]. Ignoring polarization effects, there are three contributions to the total energy of an electron in a liquid cavity, the zero point, surface tension, and pressure. These can be written as follows,

$$E = \frac{\hbar^2}{8mR^2} + 4\pi\alpha R^2 + \frac{4}{3} \pi R^3 P,$$  \hspace{1cm} (1.1)

where $m$ is the electron mass, $R$ is the cavity radius, $\alpha$ is the surface tension, and $P$ is the liquid pressure. Minimizing for the energy results in an equilibrium radius of $R_o = 19$ Å and energy $E_o = 0.21$ eV. This localized state, commonly referred to as a “bubble”, is thus energetically favorable relative to that of the free electron in uniform bulk liquid helium ($E > 1eV$).

This was not the end of the story, however. Soon after, Doake and Gribbon [1] performed mobility measurements using a triode structure, using either a Po$^{210}$ or
Am$^{241}$ source, a grid, and a collector. A constant field swept charges from the source towards the grid. They employed the Cunsolo method [20], in which the field between the grid and collector is varied as a square wave. By varying the frequency at constant amplitude and measuring the time averaged current on the collector, the minimum time necessary for the charges to travel the spacing distance can be determined, and thus the mobility. At sufficiently high values of the field between the source and grid, they observed an additional charge carrier. Their famous plot, reproduced here in Figure 1.2 shows measurements of the ion mobility as a function of square wave amplitude (i.e. drift field). These results reveal that the new charge carrier, coined the 'fast ion', travels at a velocity about six times that of the normal electron bubble in low drift fields. The mobility of the normal bubble collapses around 500 V/cm, the onset point for vortex nucleation and subsequent self trapping. The fast ion, in contrast, suffers no such collapse. To date there have been no experiments showing the existence of a fast ion critical velocity for vortex nucleation.
Ihas and Sanders quickly expanded on these results. In 1971, they conducted an experiment with a 2.6 cm long drift field, using either a tritium $\beta$ source or a needle and perforated metal plate [2, 21]. In the case of the needle, voltages were applied such that a glowing discharge was present over the liquid surface. They detected not only the fast ion and normal ion, but two additional negative charge carriers of intermediate mobilities. A subsequent experiment in 1974, utilizing the same two sources but a longer 6.5 cm drift field, revealed another nine “exotic” charge carriers with intermediate mobilities [22]. A sample trace of their data is reproduced in Fig. 1.3.

Around a decade later another research team, Eden and McClintock, designed a time of flight experiment similar to Ihas and Sanders’, utilizing a 1.0 cm drift field and a glow discharge above the liquid surface. The short time of flight distance allowed for mobility measurements at high drift fields up to $5 \times 10^3$ V/cm. They observed the fast ion, normal ion, and three of the exotic ions [23]. Additional observations revealed that unlike the fast ion, the exotic ions clearly have their high field mobilities limited by vortex nucleation, as shown on the right in Fig. 1.4. A sample trace from their data taken at $T = 1.03$ K is shown on the left of Fig. 1.4.

Most recently, Wei et al. performed a series of experiments in which they observed...
Figure 1.4: (Left) Sample traces of data by Eden and McClintock [3] (Right) Mobility of the fast, exotic, and normal ions as a function of drift field [3]

the fast ion, normal ion, and 18 exotic ions [4]. These 18 ions include those 11 that have been previously observed. Their experiment used a 6.15 cm long drift field, and a glow discharge source consisting of a sharp needle above a perforated metal plate. Both tungsten and carbon nanotube needles were used. The mobilities of the exotic ions were measured over a temperature range $T = 0.991 \text{ K}$ to $T = 1.16 \text{ K}$.

Here follows a brief overview of this thesis. In Chapter 2, I will discuss the design and basic operation of the cryostat used to cool and maintain the experiment. In Chapter 3, I provide an overview of the design of the experimental apparatus, the basic theory behind the time of flight method utilized, and the data acquisition pipeline. Chapter 4 is a detailed summary of an experiment in which the optical intensity threshold for dielectric breakdown in liquid helium was measured. This resulted in novel measurements of the way in which laser pulses modify the dielectric breakdown threshold at future times.

Finally, in Chapter 5, I detail three results from experiments in which ions were generated using an Am$^{241}$ $\alpha$-source. Their mobilities were then determined using the time of flight method in a 6.15 cm long drift field. First, a measurement of the
variation of the fast ion mobility with bulk pressure, and the implications of these results for the overall structure and origin of the fast ion. Second, I discuss new measurements of the previously observed continuous #2 background present in the exotic ion measurements of Wei et al. [8]. I will show that this background can be consistently interpreted as the partial transmission and trapping of the electron wave function in the bulk helium liquid. Third, I will discuss the observation of two previously unobserved positive charge carriers, with mobility close to that of the normal He⁺ snowball.
Chapter 2

Cryostat

I will limit discussion of the cryostat used in these experiment, since the design and construction of it has been described in great detail by Dr. Wanchun Wei in his thesis [5], and in some additional detail by Dr. Zhuolin Xie in her own [9].

The cryostat has three different temperature regions, the outermost part at room temperature, the 77 K shield in the middle, and finally the innermost 4 K shield. The experimental cell is located in the center of the vacuum space. A cross section of the cryostat is shown in Figure 2.1. The 4 K and 77 K regions are temperature maintained by the liquid helium and nitrogen baths, respectively. The combination of radiation shielding and high vacuum ($< 10^{-5}$ Torr) ensures a slow LHe and LN2 boiling rate. Typically helium transfers only need to be performed once every 36 hours, and nitrogen transfers every 8 hours. The temperature and helium level of the experimental cell are maintained independently (described in more detail below), so a great advantage of this design is that cryogen transfers do not affect ongoing experiments. This is in contrast to the previous experiments of Ihas and Sanders [2,21,22] and by Eden and McClintock [3,23,24]. They used simple dewars, in which the entire experimental cell was immersed in liquid helium. Their apparatus had the advantage of large cooling powers and the convenience of shorter cool down times,
however, there are a number of disadvantages of these types of systems. Typically, there is far less heat shielding, resulting in higher LHe usage. The immersion of the cell in the LHe bath makes the temperature harder to finely control, making temperature sensitive measurements quite difficult. Additionally, this immersion of the cell means that helium transfers result in the experimental conditions being modified. This is particularly important to avoid in exotic ion experiments, since the production conditions are notoriously fickle and not well understood. The smaller bath size and faster boiling rate also resulted in the need for far more regular LHe transfers. Finally, in these early experiments the nitrogen bath obstructed the helium bath, and thus made it hard to view the experimental cell. In our cryostat there are sets of four windows, all concentrically aligned. Each set is 90 degrees apart, and consists of windows on each of the three tails and the cell. Since these windows are below the nitrogen and helium baths, the view of the cell is unobstructed, opening up a variety of experimental possibilities. All this is to say, the complexity of the larger cryostat system is worthwhile since it avoids the issues associated with a simpler one.

The experimental cell is a stainless steel cylinder, 4 3/8” tall and 2 1/2” in diameter. As mentioned above, there are four windows on the cell. Each is 3/4” in diameter, providing good views of the inside of the chamber, and access for lasers and other light sources. The cell is filled with liquid helium using two independent filling lines. To reduce impurities, only 99.999% pure helium gas cylinders are used. The helium gas travels, via copper piping, from outside the cryostat through a nitrogen cold trap and then through the helium bath to transfer out most of the heat. After passing through the helium bath, the helium liquid passes into narrow stainless steel capillaries. These capillaries are connected to heat exchangers at the 4 K ring and the 1 K pot to provide addition cooling. Finally, the capillaries terminate at the cell, where they are brazed onto stainless steel flanges, which are indium sealed on the side of the cell.
Figure 2.1: Cross section of the cryostat showing the main cryogenic systems, from W. Wei’s thesis [5].
A bottom plate with a series of electrical feedthroughs is indium sealed to the main portion of the cell. There are three single pin high voltage feedthroughs, typically used for the discharge source (S), the first mesh grid (G0), and the second mesh grid (G1). In addition there are two low voltage feedthroughs, with five pins each. One is used for the third mesh grid (G2) and the four field homogenizers (H1, H2, H3, H4). The other is used for the collector (C) and Frisch grid (F). This leaves a few utility pins for additional components, providing experimental flexibility.

Cooling of the cell is provided by the 1 K pot, which is run continuously while the cryostat is at low temperature. The 1 K pot is a 221 cm$^3$ cylindrical cell filled with helium from the bath. The liquid is first cleaned by a fine mesh filter at the bottom of the bath, and then fed through a stainless steel capillary. It is pumped by an Edwards 1250 Roots pump backed by an Alcatel 2063H rotary pump. A number of features are included to maximize the cooling rate. The filling capillary is thermally anchored to the pumping line, so that the outgoing cold vapor transfers heat from the incoming $\sim 3$ K liquid. The bottom of the 1 K pot and top of the experimental cell are actually the same oxygen free copper (OFC) surface. The temperature difference across the interface arises from the thermal resistance of the metal itself and the Kapitza resistance of the two helium to metal interfaces. The thermal resistance of the copper, however, is negligible compared to the Kapitza resistance. 1/8" x 1/8" grooves are cut into both sides of the OFC interface, perpendicular to each other, in order to maximize the surface area and thus minimize the Kapitza resistance. The net result is a 1 K pot which can maintain a temperature difference of only 8 mK at 1 K under a 50 mW heat flux.

Temperature monitoring during the cooling down process is provided by three silicon diode thermometers, at the helium bath, the 4 K ring, and the experimental cell. These are monitored by Lakeshore 330 and 331 temperature monitors. During normal operation two germanium thermometers are used, one at the 1 K pot pumping line,
and the other on the experimental cell flange. These are monitored by a Conductus LTC-20 temperature controller. The 1K pot is heated by a 3.69 kΩ resistor mounted to the 1 K pot. By adjusting the voltage to the 1 K heater while maintaining a constant cooling power, we can adjust the cell temperature. Adjustment of the heater is done automatically by the LTC-20. With this arrangement the temperature can be controlled to 1 mK precision at reasonably static heat fluxes (in most cases from the ion discharge).
Chapter 3

The Experimental Cell and Time of Flight Method

Charged objects injected into liquid helium quickly lose any kinetic energy via collisions, transferring from free states into larger composite ones. These generally have two forms. There are bubbles, formed by loosely bound charges, in which the kinetic energy of the core is large enough to force neutral helium away, forming a cavity. There are also objects referred to as snowballs, formed from tightly bound positive charges. The charge being tightly bound allows the polarization interaction with neutral helium atoms to dominate, such that these atoms are drawn in by electrostriction and form a high density solid cluster. This is far from a complete picture, the exact form of charged objects in the helium is determined by the geometry of the object’s wave function. Critically though, in all cases these objects are quite large compared to the spacing between helium atoms, on the order of 10 Å. The large size generates a significant drag force. When a field is applied the terminal velocity is easily measurable in the lab. The mobility of the ions is defined as the ratio of average velocity $v$ to electric field $E$. 
\[ \mu = \frac{v}{E} \]  

This has led to decades of experiments which largely utilize the same basic apparatus, i.e. a source of charged particles, a gate, a drift field, and a collector. The gate controls the flow of charges, allowing a small number of ions from the discharge to pass through at a chosen time. The drift field is a static electric field through which the objects experience a near constant force. After traveling the full length of the drift field, the objects are then accumulated on the collector and measured as a time resolved current. From the dependence of each ion’s mobility on the state of the liquid (pressure, temperature, etc.), properties like the size and structure can be inferred.

### 3.1 Time of Flight Apparatus

In this section I will provide an overview of the apparatus used to conduct the three experiments described in Chapters 5. It was modified for the experiment described in Chapter 4, but those changes will be discussed there. The initial version of this apparatus was designed and constructed by W. Wei [5], and used in a series of experiments studying the fast and exotic ions [8, 25, 26, 27].

A cross section of our experimental cell is shown in Fig. 3.1. Here we can see the main components of the experiment, discussed in detail in the following sections. From top to bottom, there is the discharge region, consisting of a radioactive \( \alpha \)-source (S) and mesh grid (G0). Below that is the gate region, consisting of mesh grids G1 and G2. Voltages on G1 and G2 are used to control the flow of charges into the drift field region below, consisting of G2 and field homogenizers H1-H4. Finally, in the collection region, there is a Frisch grid (F) for screening field lines and a brass disc (C) for collecting charges. These are the only parts of the experiment directly
related to producing, moving, and measuring the ions. The outer stainless steel cell, electrical feedthroughs, etc. are not shown.

3.1.1 Source

In our experiments, the arrangement of the sharp discharge tips and the perforated plate (P) used by Wei et al. were replaced by an Am$^{241}$ α-source (S) and mesh grid (G0). Tungsten tips were previously used to generate exotic ions by Ihas and Sanders and Eden and McClintock, and both tungsten and carbon nanotube tips were used by Wei et al. These tips are capable of producing large discharge currents, however they are unreliable. During Wei’s experiments the signal from the tungsten tips would often degrade quickly. The carbon nanotube tips produced exotic ion signals as large as 10 pA in some cases, but they proved difficult to consistently manufacture and degraded during experiments in the same manner.

From 2015-2016 we performed a series of experiments in which we tried to produce a more consistent carbon nanotube based signal. To produce CNT tips, Wei cut sections of thin stainless steel wire, coated it in silver epoxy, and then rolled the epoxy covered tips in a mixture of acetone and CNTs. The tips were left for at least a few days to allow the epoxy to cure. Initially we used the same method, but attempted to enhance quality control by throwing away tips that had an uneven CNT coating, bald spots, or those in which the epoxy did not adhere the CNTs to the wire well. Efforts to observe the exotic ions were still inconsistent, rarely producing exotic ions, and if they did only for brief periods before failing.

Another attempted solution involved modifying the discharge geometry. The plate was redesigned to be more like that of Eden and McClintock. Tungsten tips were soldered to the perforations in the plate such that their sharp ends directly faced the CNT tips. This must have produced a much stronger discharge field by nature of the high electric fields around sharp points, but it still failed to produce strong exotic ion
Figure 3.1: Cross section of the experimental cell. S is the Am$^{241}$ α-source, G0, G1, and G2 the mesh grids, H1-H4 the homogenizer discs, F the Frisch grid, and C the collector. $R_c$ is the collector load resistor.
signals for more than a few hours.

Finally, we hypothesized that the quick degradation of the tips was from the CNTs dulling. The tips becoming dull would increase the field strength necessary to produce a plasma. The tips were tested at room temperature in helium gas, with the pressure chosen so that the density was approximately the same as the density of T = 1.0 K helium at SVP. We observed that the plasma would often preferentially form between certain tips and not others. In some experiments a plasma would form evenly in the entire plate/tip region, but after some hours abruptly change states. At these later times, the plasma formed only between the plate and one tip. In response, we manufactured a flat stainless steel disc, to which we then applied a solution of high quality CNTs (courtesy of the Xu group at Brown University) and allowed to dry. This formed a more uniform surface, and thus a more uniform discharge field. Although this produced a long lasting plasma in room temperature tests, it failed to produce exotic ions at low temperature.

Eventually we decided to test out entirely different types of sources. One new approach that we tested for generating ions was using a laser to induce dielectric breakdown of the helium liquid. A high intensity laser was aimed at the cell window, and a lens placed inside the cell focused the beam onto a point above the drift field and along the central axis of the cell. When the intensity at the focal point was high enough, an ionization cascade was induced, resulting in large numbers of ions being produced. Although this approach failed to work as a time of flight experiment, it did lead to a series of experiments studying the breakdown threshold of liquid helium. The results of these experiments will be discussed in Chapter 4.

As a result of these issues, we decided to switch to a 300 $\mu$C Am$^{241}$ source. The Eckert & Zeigler Am$^{241}$ source has a 5 mm platinum foil onto which the active Americium is electroplated. The $\alpha$-particles are ejected with an energy of 5.5 MeV. The source is mounted in a 1.6 cm diameter brass disc to which a wire is attached via
Figure 3.2: Cross section of the S-G0 nylon spacer

2-56 screw. It is separated by a 1.75 mm thick nylon disc with 5 mm inner diameter. The design of this spacer is shown in Fig. 3.2.

3.1.2 Gate and Drift Field

The grids, G0, G1, and G2, consist of 0.5 mm thick nickel discs with 8 mm center holes, onto which wire grids are spot welded. The wire grids are made of nickel, with 39 micron diameter wires and 28 wires per centimeter spacing. After the spot welding, the grids were sputtered with gold and palladium. The flow of current into the drift field below is controlled by creating a gate between G1 and G2. G1 is normally held slightly less negative than G2 to produce a reverse field. To produce a pulse of ions, a negative voltage pulse is delivered to G1 (and G0) so that G1 is slightly more negative than G2. This allows a thin slab of ions to pass into the drift field. G0 and G1 are separated by a stack consisting of a brass electrode, a nylon spacer, and another identical brass electrode. Together these three parts separate G0 and G1 by 5.6 mm. G1 and G2 are separated by a 1.5 mm thick nylon spacer.
The drift field consists of G2 and four stacked homogenizer units. Each homogenizer unit is formed by stacking a 5 mm thick brass electrode, a 0.5 mm thick nickel disc with 1.27 cm inner diameter (the homogenizer disc), another identical 5 mm brass electrode, and finally a 1.5 mm thick nylon spacer, making each unit of the drift field 1.23 cm thick and the total drift field 6.15 cm long. Rather than apply voltages individually to the homogenizer discs, a resistor chain is formed inside the cryostat by five $1 \, M\Omega$ resistors to ground, so that their voltages are evenly spaced and set by G2, ensuring good field uniformity.

3.1.3 Collector

The Frisch grid sits at the bottom of the drift field. It is constructed from a 0.5 mm thick nickel disc, identical to the grids described above except with a larger 1.27 cm inner diameter. Without the Frisch grid shielding the collector, approaching charges would induce an image charge on the collector, and thus an unwanted signal. The collector sits underneath the Frisch grid and is made from a 1.27 cm diameter brass disc. The Frisch grid and collector are separated by a 1.5 mm nylon spacer. It sits inside a nylon ring which electrically isolates it from the bottom of the cell. Charges absorbed on the collector are passed via miniature coaxial cable to a 2.5 $M\Omega$ load resistor. This resistor is heat sunk to the side of the experimental cell to reduce thermal noise in the signal. Another miniature coaxial cable runs from the load resistor to the outer jacket of the cryostat. An approximately 2” BNC cable from the outside of the cryostat runs to a Stanford Research Systems SR560 unit, which acts as both a preamplifier and a low pass filter. Typical amplifications used are of the order $10^3$ and the low pass filter is at 30 kHz with a -12 dB/octave roll-off. Finally, the amplified signal output of the preamplifier is fed to a National Instruments BNC-2120 connector block and National Instruments PCI-E6361 data acquisition card.

This arrangement ensures the shortest possible signal path from the cryostat to
the preamplifier. This, combined with the use of miniature coaxial cable, produces a low total capacitance, with a measured RC time constant of only $\sim 170 \mu s$. This is essential both for limiting the effects of electrical pickup from the gate pulse, as well as for resolving the smaller exotic ion signals.

3.1.4 Electronics

The essential circuitry is diagrammed in Figure 3.3. The source S is kept at voltage by a Stanford Research Systems PS350 high voltage power supply, and current limited by a resistance of 12 MΩ. G0 and G1 are both current limited by 1.2 MΩ. 50 µF and 0.03 µF capacitors in series pass the gate pulse to G0 and G1. G2 and F are each protected by 50 kΩ resistors, and 0.005 µF of capacitance to ground. These limit high frequency voltage fluctuations that would otherwise induce noise on the collector. Because of these protection circuits, the voltage on the power supplies is not equal to the voltage on the electrodes. The voltages on S, G0, G1, and G2 are measured by digital voltmeters connected through 20 MΩ, 1000:1 voltage dividers. F is monitored with a Hewlett Packard digital multi-meter.

Each experimental trace is driven by a Berkeley Nucleonics pulse generator. It outputs a TTL pulse, typically every 100 ms. The TTL pulse serves as a trigger for the DAQ card to begin recording, as well as to drive the General Radio 1217 pulse generator. The pulse generator outputs a square wave pulse, typically $\pm 40V$, to G0 and G1.

3.1.5 Signal Acquisition

The PCI-e DAQ card acts as an analog to digital converter. Each trace, typically 75 ms long at a 40,000 Hz sampling rate, is recorded by the computer. Because each individual trace has low signal to noise, we record many traces (most often 3,000) and average them. The 170 µs time constant of the collection circuit produces high
Figure 3.3: Circuit diagram of the time of flight circuit
frequency noise in the raw signal. Wei noted in his thesis that thermal noise and noise from the preamplifier were insufficient to explain the observed noise \[5\]. Instead he attributed the noise to electrical pickup and pump vibrations. To remove the noise, a smoothing process is employed by convolving the averaged signal with the square of a cosine. The argument of the cosine goes over one half cycle and the frequency is variable (set manually to adjust the degree of smoothing). Figure 3.4 contains an individual trace (top) and then the result of a 3,000 trace averaging and smoothing. After averaging and smoothing, the appearance of the normal electron bubble is clear (denoted by “NB” in Fig. 3.4).

3.1.6 Interpreting the Traces

Qualitatively, a single run (trace) of the experiment can be broken up as follows:

- Gate is closed, charges produced by S-G0 field are either absorbed onto G1 or remain in the liquid nearby.
- Trigger sent, G0 and G1 pulsed, gate opened, computer begins recording
- Ions travel towards the drift field region at a speed \(\mu E_{G1-G2}\)
- Gate closes after a time \(\Delta t_{gate}\). Ions that traveled past G2 before the gate closes continue downward
- The slab of ions travels from G2 to F at a velocity \(\mu E_d\). Ions with different mobilities separate into their own slabs during this time.
- As the front of each slab passes through F, the voltage on \(R_C\) begins to rise with a time constant determined by the capacitance of the electrodes, cables, and input of the preamplifier.
Figure 3.4: (Top) Single experimental trace, (Bottom) Averaging of 3000 traces, fast ion signal becomes clear
A straightforward analysis of the collection circuit (including the Frisch grid) shows that the current induced by a thin layer of surface charge density $\sigma$ and area $A$ on the collector depends only on the velocity of the charges,

$$I = \frac{\sigma A}{d_{FC}} v_{FC}.$$  \hfill (3.2)

Here $d_{FC}$ is the spacing between the Frisch grid and collector, and $v_{FC}$ is the velocity a particular ion travels between them. Let us now consider an ion slab of finite thickness. As the front edge of the slab passes the Frisch grid, the amount of charge between F and C will increase at a constant rate, and thus so will the charge induced on C. By the same argument, the charge will reach a maximum when the rear of the ion slab passes through F and the charge in the F-C region is maximal. Finally, the induced pulse will fall off at the same rate as the initial rise.

Now we consider how the gate pulse, of width $\Delta t_{gate}$, affects the signal. The time from the beginning of a pulse to its peak is approximately the effective gate width for that particular ion, $\Delta t_{eff}^{gate}$. $\Delta t_{eff}^{gate}$ is less than $\Delta t_{gate}$ because it takes some time for the front of the ion slab to pass from G1 to G2, but in almost all cases this transit time is so fast compared to $\Delta t_{gate}$ that we effectively have $\Delta t_{eff}^{gate} = \Delta t_{gate}$. Thus, the time that passes from the beginning of the gate pulse to the arrival of an ion pulse is approximately the same as the time from the end of the gate pulse to the peak of the ion pulse, i.e. $t_{arrival} = t_{peak} - \Delta t_{gate}$.

To simplify the calculation of the mobility $\mu$ further, we approximate the drift field as $E_d = \frac{V_{G2} - V_F}{L_d}$. Together, this gives,

$$\mu = \frac{L_d^2}{(V_{G2} - V_F)(t_{peak} - \Delta t_{gate})}.$$  \hfill (3.3)

And so by identifying the ion peaks and their associated maxima, the ion mobility can be easily calculated.
Chapter 4

Laser Induced Breakdown

Measurements in Liquid Helium

With the introduction of high powered lasers in the mid twentieth century, it became possible to produce dielectric breakdown in a variety of materials. Besides illuminating the underlying physics of the ionization process, such studies hold value in applications such as the detection of impurities and understanding the damage thresholds of various materials. Of particular interest are past experiments in which lasers were used to successfully produce break downs in liquid helium. Using these past studies as inspiration, we set out to further study the properties of exotic ions in liquid helium, by devising an experiment to induce optical breakdown in the liquid and detect any bubbles formed as a result. Although this experiment proved unsuccessful, it led to a series of additional measurements of the breakdown process in liquid helium. In this experiment we use visible and near visible pulsed laser emission to examine the breakdown threshold properties of helium in the superfluid state, including the role of laser energy, temperature, time between pulses, as well as previous pulses and breakdowns.

The results detailed in this chapter were previously published in 2017 in the Jour-
4.1 Summary of Previous Results

There have been a number of studies of the breakdown threshold in helium gas [29, 30, 31, 32], but studies of the breakdown threshold in liquid helium are more limited. Winterling, Heinicke, and Dransfeld [6] used a 25 ps, 694 nm Q-switched ruby laser to measure the breakdown threshold from 1.1 K - 4.0 K. Pulses were fired at a time interval of approximately 10 minutes [33]. They found that the breakdown threshold varied between $\sim 10^{10}$ and $\sim 10^{11}$ W cm$^{-2}$, dropping an order of magnitude between 1.5 K and $T_\lambda$. At temperatures above this region, they found that the breakdown threshold does not depend on temperature. Benderskii et al. [34], using a 790 nm laser with 80 fs pulses, measured a threshold of $\sim 5 \times 10^{13}$ W cm$^{-2}$ between $T = 1.45$ K and $T = 4.0$ K. Gao et al. [35] found a threshold of $\sim 10^{14}$ W cm$^{-2}$ using 35 ps, 800 nm pulses at $T = 1.83$ K.

Detailed discussions of the theory of optical breakdown have been provided in papers first by Keldysh [36], and then by Zel’dovich and Raizer [37]. Keldysh described the optical process as a dynamical one, split into two regimes. In the first regime, tunneling is the dominant process for ionization. For a helium atom of ionization potential $U_I$, subject to a laser field of intensity $E$, an electron sees an effective tunneling barrier length

$$l = \frac{U_I}{eE}$$ (4.1)

Their average velocity is determined by $U_I$, $v = (2 U_I/m)^{1/2}$, where $m$ is the electron mass. These two parameters define a tunneling frequency,

$$\omega_t = \frac{eE}{\sqrt{2m U_I}}$$ (4.2)
If the laser frequency is higher than $\omega_t$, the tunneling probability decreases, since the electron can’t travel the tunneling distance $l$ before the field changes sign.

In the second regime, dominated by frequencies and field strength combinations where tunneling is typically forbidden, ionization proceeds by simultaneous multiphoton absorption. Keldysh combined these two regimes within the same framework, showing that they were actually limiting cases of the same process. Within this framework, the dominant ionization regime is determined by the Keldysh parameter $\gamma$, defined as the ratio of the optical frequency to the tunneling frequency, $\gamma << 1$ for tunneling and $\gamma >> 1$ for multiphoton.

$$\gamma = \frac{\omega}{\omega_t}$$ (4.3)

Zel’dovich and Raizer [37] expanded upon this by noting that in helium gas breakdown experiments by Meyerand et al. [38], breakdown was occurring at laser fields far lower than that predicted by the Keldysh theory. Charges collected from the breakdown region outnumbered, by many orders of magnitude, what was expected from the theory. Zel’dovich and Raizer argued instead that the breakdown proceeds through a “cascade process” [39]. A few “priming” electrons, present in the focal region, are accelerated in the laser field. Although free electrons cannot absorb photons directly, they can absorb photons during an atomic scattering interaction. The electrons gain and lose energy through these scattering processes, effectively changing energy randomly in time, or diffusing along the energy axis. The electrons undergo this process until they have accumulated energy greater than the ionization potential $U_I$, at which point it becomes very likely that the electron will ionize a nearby atom, lose most of its energy, and begin the process over. The newly liberated electron does the same, resulting in successive generations and an exponential increase in the rate of ionizations. In most cases, the frequency of collisions, $\omega_{coll}$, is less than the light frequency $\omega_{light}$, but if the density is sufficiently high it may approach or exceed it.
This energy gain process is hindered, however, by the fact that electrons can lose their energy to intermediate energy levels (excited states), $E_n$, making it difficult for any electrons to acquire energy $U_1$. Additionally, electrons can diffuse out of the focal region. Zel’dovich and Raizer proposed impurities and rare direct ionization events as possible sources for these “priming” electrons.

As mentioned before, Winterling et al. [6] observed a sharp drop in the breakdown threshold for nanosecond pulses beginning at a temperature of $T = 1.5$ K. At low temperature the density of the liquid changes by less than 1% from $T = 1.5$ K to $T_\lambda$, so $\omega_{\text{coll}}$ varies very little. Thus, in terms of the Zel’dovich and Raizer theory, such a sharp change in the breakdown threshold is unexpected. Winterling et al. proposed that the collisions of the priming electrons were dominated by scattering with rotons, rather than with helium atoms. They argued that because the roton density decreases below $T_\lambda$, $\omega_{\text{coll}}$ would also decrease and the breakdown threshold would thus increase.

To explain the threshold being independent of temperature below $T = 1.5$ K, they argued that the spontaneous generation of rotons by electrons would keep the roton density from dropping further. Silver, Hernandez, and Onn [40], however, argued that this explanation was incomplete, citing previous measurements [41] of the electron scattering rate that show little variation from $T = 1.35$ K to $T = 4.2$ K. In a separate paper Hernandez and Silver [42] proposed that the temperature dependence of the breakdown threshold resulted from the role electron bubbles play. At higher temperatures the incipient bubbles would be readily destroyed by roton interactions, but as the roton density drops they would favor stabilizing.

In a private communication with Hunklinger and Leiderer, Winterling and Heinicke [33] discuss observing a time dependence in their measurement of the threshold. Initially producing a breakdown in the liquid required a laser intensity an order of magnitude higher than that required to produce breakdown again a short time later. Abrikosova and Bochkova [43] reported observing a similar time dependence. This
time dependence suggests that a source of electrons remains in the focal region after the first breakdown, explaining the lower breakdown threshold. A number of experiments have been performed to understand the origin of such initial “priming” electrons in producing breakdown in the liquid helium. Winterling et al. [6] tested whether free electrons remain in the focal region after the first laser pulse by applying a 1.5 kV cm\(^{-1}\) electric field. The application of the field was found to have no effect on the breakdown threshold, however. Hunklinger and Leiderer [44] used a hot cathode to inject free electrons (\(\sim 10^3\)) into the focal region, but also saw no change in the threshold intensity. These experiments both indicate electrons ’leftover’ from previous pulses are not responsible for lowering the threshold on subsequent pulses. 

Abrikosova and Bochkova [43, 45, 46] considered the possibility that the electrons needed to initiate a breakdown originate from impurities of low ionization energy in the liquid. The impurities could be ionized by the laser either indirectly, by heating, or directly, by photo-emission. If these impurities resulted in large numbers of free electrons, then the injection of a small number of electrons, like that by Hunklinger and Leiderer, would have no observable effect on the threshold intensity. They tested this hypothesis in two experiments. In the first, Abrikosova and Bochkova [43] added small amounts of hydrogen into the liquid helium. With condensed hydrogen in the liquid, they found a lower and consistent breakdown threshold. In the second experiment, Abrikosova and Skyrpnik [45] cleaned the focal region of impurities, by pushing a fine mesh filter (“Petryanov filter”) through the liquid. After this cleaning process no breakdown could be detected, up to the maximum intensity of the laser, \(\sim 6\times 10^{10} \text{ W cm}^{-2}\).

The experiments of Abrikosova and Bochkova suggest that impurities are the source of the initial electrons, but this by itself still fails to explain the time and temperature dependence of the threshold. Hunklinger and Leiderer [44] proposed that the sharp change in threshold around \(T_{\lambda}\) might arise from sedimentation in the
liquid. Below $T_\lambda$, in the superfluid, impurities would sediment out faster than in the normal fluid, resulting in less impurities at the focal region. This description seems insufficient, however. At higher temperatures, there would be large impurity densities, the probability of breakdown would very nearly be 1, and the threshold would thus be a lower value, $I_l$. As the impurity density gets low compared to the size of the focal region, the breakdown should depend on the probability $p$ of finding an impurity in the focal region. If there are no impurities present, a higher threshold value $I_h$ would be measured. We would then expect the probability of a threshold measurement resulting in $I_h$ to be $1 - p$, and $p$ for $I_l$. This contradicts the results of Winterling et al., who observed a continuous change in the threshold.

Another proposal for the origin of the breakdown threshold, made by Benderskii et al. [34], is neutral helium dimers [47]. Neutral helium dimers have a long lived triplet state, $\text{He}_2^* (3a)$. These excited dimers, or excimers, have a much lower ionization potential of only 4.3 eV, providing a much more ready source of electrons. In their experiment, Benderskii et al. used 790 nm, 80 fs, 500 Hz laser pulses at intensities of $10^{13}$ W cm$^{-2}$. By using a pump-probe scheme, they performed time resolved measurements of the excimer density between pulses, showing conclusively that $\text{He}_2^*$ molecules were produced during breakdown and decayed bimolecularly afterwards. This explanation is seemingly consistent with the experiments above. The neutral dimers would remain unperturbed by an electric field like that introduced by Winterling et al. [6]. They more readily provide free electrons, so given a sufficient density of dimers the null result of Hunklinger and Leiderer [44] in injecting electrons would be expected. Finally, the Petryanov filter used by Abrikosova and Skyrpnik [45] may have filtered the dimers out of the focal region, or destroyed them since the dimers are only very loosely bound by the van der Waals force. Estimates of the dimer population at the focal region are difficult to make. The dimers have a lifetime of $\tau = 13$ s [48], close to the typical repetition rates of the laser. They destroy each
other in bimolecular Penning ionization processes at a rate proportional to the square of their density \[49\]. They will also naturally diffuse through the liquid.

### 4.2 Apparatus

The source of pulses in our experiment is an EKSPLA NT342/1 ND:Yag laser \[50\] lent to us by the National High Magnetic Field Laboratory. Light from an external flash lamp, with adjustable intensity, is directed via an optical fiber to the ND:Yag rod. Stimulated by the flash lamp, the laser rod produces a 1064 nm (1.16 eV) beam, which is directed toward nonlinear crystal frequency doublers, resulting in additional 532 nm and 355 nm pulses. The maximum pulse energy is approximately 250 mJ (1064 nm), 50 mJ (532 nm) and 15 mJ (355 nm). The pulse duration is 3 to 5 ns. The repetition rate of the laser is controllable and ranges from 0.1 seconds to 10 seconds.

The laser pulse is focused with a Thor Labs aspherical \( f = 11.0 \) mm lens, placed inside the experimental cell. The lens is placed in a 1/4 inch channel machined through the side of a thick nylon disc. This channel allows light to escape from the other side, and thus minimize heating of the liquid. An identical channel is machined perpendicularly, to allow an unobstructed view of the focal region. Electrodes are located directly above and below the lens. Further below is the time of flight apparatus, identical to that designed by W. Wei \textit{et al}. \[26\] A cross section of the full cell design is pictured in Figure 4.1. Outside the cell, 1064 nm\( / 532 \) nm dielectric mirrors from Thor Labs are used to direct the pulse into the cell with minimal intensity loss.

An estimate of the size of the focal region was made at room temperature. The laser pulse was directed onto a piece of scotch tape placed on a glass plate. A picture of the focus (Figure 4.2) is taken by first focusing a digital camera on the scotch tape, and then adjusting the position of the lens so that the focus of the lens is also on the
Figure 4.1: Cross section of the apparatus for laser induced breakdown experiments. The $f = 11$ mm lens (green) is held in place in a 1/4 inch diameter cylindrical channel. This channel runs perpendicular to the drift field axis, and was machined into a nylon disc. The time of flight apparatus from previous electron bubble experiments sits below this nylon disc.
scotch tape. The resultant image is fit with a Gaussian profile, and the full width at half max, 50 µm, is taken to be the focal diameter. Because we lacked an infrared CCD, this measurement was only performed with the 532 nm beam.

To avoid any large uncertainties in the pulse energy, either due to the warm up time of the laser or from the analog controls on the pump beam, an external Ophir power meter was used anytime the intensity was adjusted.

4.3 Results

4.3.1 An Experiment to Produce Electron Bubbles

The threshold experiments detailed in this chapter grew out of an initial attempt to produce electron bubbles from a laser induced breakdown. Both tungsten and carbon nanotube tips, although greatly successful in some experiments, have proved frustratingly inconsistent in producing exotic ions for study. The experimental apparatus was designed around maintaining the existing drift field and collector structure used in the mobility experiments, and making the minimal addition of an electrode directly
above the breakdown region so that any electrons liberated by the breakdown could be directed into the drift field.

Though the focused pulse successfully initiated a breakdown of the liquid, an ion signal from the breakdown could not be successfully detected. For a period of tens of milliseconds immediately after the pulse enters the cell, the signal at the collector becomes dominated by high frequency, high intensity noise that is much larger than the typical ion signal. A representative signal of this noise is shown in Figure 4.3. The noise is believed to be from sound pulses produced by the sudden heating of liquid at the focal point. Indeed, even at laser intensities close to the threshold for producing a breakdown, a 'pinging' sound could be heard from outside the cryostat. At higher laser intensities this noise became quite loud. We did not keep the intensity high for very long for fear of producing sound pulses which might have damaged the experiment.

Additionally, we searched for currents produced by laser pulses which are below
the intensity needed to produce an avalanche breakdown. The collector noise was reduced as compared to the higher laser intensity, but noise on the order of the typical exotic ion signals still remained. No normal ion or exotic ions signals were discernible. This noise level remained unchanged even when the laser was blocked, so we believe it was the product of electrical pickup from the flash lamp circuitry.

4.3.2 Measurement of Breakdown Thresholds

We began by taking a preliminary measurement of the temperature dependence of the threshold intensity in liquid helium. However, it quickly became apparent while doing this that when a breakdown is produced in the liquid, the probability of a breakdown occurring on a subsequent pulse was greatly increased. At a constant pulse intensity, many consecutive pulses would not produce a breakdown, but as soon as a breakdown did occur, almost all subsequent pulses also did. Additionally, even at a constant intensity, the number of pulses needed to cause a breakdown after the laser was turned on varied dramatically. Such results imply a strong time dependence of the breakdown threshold. As such, we decided it was critical to measure the threshold in a variety of ways, and clearly define the measurement processes used.

4.3.2.1 Measurement of the Threshold for Initial Breakdown, $I_1$

In the first series of measurements, we chose to examine the threshold intensity of the liquid, at 1064 nm, when it had had a long time to recover from the effect of any previous pulses. The liquid was left undisturbed for at least 60 seconds, and then two laser pulses were sent into the cell two seconds apart. The intensity at which at least one of these two pulses produces a breakdown at least 50% of the time is defined as $I_1$. The results for $I_1$ from $T = 1.1 \text{ K}$ to $T = 3.0 \text{ K}$ are in Figure 4.4. Within the experimental errors, $I_1$ stays approximately constant at $8 \times 10^{10} \text{ W cm}^{-2}$, in contrast to the steep drop in intensity around $T_\lambda$ observed by Winterling et al.
A measurement of \( I_1 \) was also made at \( T = 1.1 \) K using 532 nm pulses. We found that \( I_1 = 6 \times 10^9 \) W cm\(^{-2} \). This is likely explained by the factor of two reduction in quanta needed for multiphoton ionization. Unfortunately, the harmonic generating crystals also produce a non-linear increase in the per pulse energy variation. Our initial goal was to repeat those measurements previously taken with the 1064 nm beam, but this variation was too large for such data to be meaningful.

### 4.3.2.2 Measurement of the Threshold to Maintain Breakdown, \( I_2 \)

As noted at the beginning of the section, it was observed that the breakdown probability is greatly increased if the previous pulse resulted in breakdown. To study this further, we set out to determine the lowest laser intensity which can maintain the breakdown condition in the liquid. For this measurement, the laser intensity was increased sufficiently to produce a breakdown, and then pulses continued to be applied every 2 seconds. Next, the laser intensity was lowered in small increments, until the intensity was sufficiently small such that no pulse produced a breakdown for at least 60 seconds. This intensity, referred to as \( I_2 \) in Figure 4.4, is approximately 4 times lower than \( I_1 \) across the temperature range. Like \( I_1 \), \( I_2 \) shows no sudden drop around \( T_\lambda \).

To further examine the role of previous breakdowns in changing the state of the liquid, we measured \( I_2 \) while varying the laser repetition rate. If we presume that the liquid does indeed “restore” after a given length of time, the most natural assumption is that \( I_2 \) will increase and approach \( I_1 \) as \( \tau \) increases. The temperature was maintained at \( T = 1.1 \) K. As shown in Figure 4.5, \( I_2 \) does indeed behave this way.

A short test similar to that of Winterling et al. \([6]\) was performed during a state of continuous breakdowns. Using the electrodes inserted above and below the focal region, fields up to the order of a kV/cm were applied. These fields had no apparent effect on ceasing the breakdown at a given threshold intensity, so we also conclude that
Figure 4.4: Laser intensity required to produce breakdown as a function of temperature. The diamonds are the results obtained by Winterling *et al.*, [6], and the crosses and squares are the results obtained in the present work. The definitions of $I_1$ and $I_2$ are given in the text.

Figure 4.5: Laser intensity $I_2$ as a function of the time $\tau$ between successive pulses. The definition of $I_2$ is given in the text.
the source of charges for breakdown is not charged particles leftover by the previous pulse.

4.3.2.3 The Effect of Laser Pulses That Do Not Produce Breakdown

$I_1$ is a measure of the initial threshold intensity at which a breakdown nearly always occurs, while $I_2$ is a measure of the lowest intensity needed to maintain the liquid condition for breakdown between subsequent pulses. The results of these measurements clearly show that a breakdown in the liquid temporarily changes the state of the liquid, such that a subsequently applied pulse is more likely to result in the liquid breaking down. As noted above, it was often the case that at a given laser intensity, many pulses could be applied to the liquid before breakdown occurred. In the next set of measurements, we set out to examine whether this process is purely random at a given intensity, or if pulses which do not produce breakdown also affect the probability for subsequent pulses.

First, the pulses were blocked from entering the cell for at least 60 seconds, then pulses were applied at a repetition period $\tau$ until a breakdown occurred. The number of pulses required was counted, and the measurement iterated 220 times. The temperature was maintained at $T = 1.1$ K, and the laser intensity at $I = 6.7 \times 10^{10}$ W cm$^{-2}$. The laser intensity was chosen by manually adjusting for a value between $I_1$ and $I_2$ at which the typical number of pulses applied before a breakdown was reasonable for counting by eye. 220 data points were collected for $\tau = 0.5$ s and 1.0 s each. These results are in Figure 4.6.

If the pulses which do not produce breakdown have no effect, the probability of producing a breakdown, $q$, should be identical for each pulse. The probability of a breakdown on the $n$-th pulse, $p(n)$, is therefore

$$p(n) = (1 - q)^{n-1}q \quad (4.4)$$
This implies that the first two moments of the distribution would be as follows:

\[ \langle n \rangle = \frac{1}{q} \]  
\[ \langle n^2 \rangle = \frac{2 - q}{q^2} \]

For the measurements at \( \tau = 0.5 \) s, \( \langle n \rangle = 17.0 \) and \( \langle n^2 \rangle = 600.7 \), which gives \( q = 0.059 \) and \( q = 0.057 \) respectively. The measurements for \( \tau = 1.0 \) s give \( q = 0.049 \) and \( q = 0.050 \). The values for \( q \) agree well in both cases, indicating no significant divergence from the distribution in Eq. 4.4 and thus that the probability of producing breakdown is constant assuming a breakdown has not occurred recently.

We can go further and ask if the \( q \) values for the two repetition rates are statistically compatible, i.e., does the probability of producing an initial breakdown depend on the repetition rate. To estimate the expected variation in experimental values of \( q \), 220 pseudo experiments are performed at a particular breakdown probability value, \( q_0 \), to form a sample distribution, and this is repeated \( 10^6 \) times. Eq. 4.6 is then used...
to calculate $q_n$ for each of the $10^6$ distributions. The distribution of $q_n$ values is fit with a Gaussian. For $q = 0.050$ and $q = 0.058$, the results of the fit are identical, with $\sigma = 0.004$. Thus, we find that the values for $q$ derived from the experiment are compatible within $1\sigma$ error bars.

### 4.3.2.4 Source of Priming Electrons

The parameters of the experiment fall strictly within the multiphoton regime of the Keldysh framework. This explanation, on its own, is insufficient however. For the 1064 nm light predominantly used, $n = 24$ photons would be required for direct multiphoton ionization of the ground state helium atoms. Yet in the focal region the ratio of photons to helium atoms is $\sim 10^{-2}$, indicating high multiplicity events would be exceedingly rare.

Helium dimers, however, have an ionization potential of only 4.3 eV, and would thus require only $n = 5$ photons. If laser pulses, causing breakdown or not, do produce dimers in the focal region, it is important to note that most would diffuse away before the next pulse. The diffusion coefficient is $\sim 0.007$ cm$^2$s$^{-1}$ at $T = 1.1$ K \cite{51}. The typical time between pulses is $\sim 1$ second, and so dimers would travel a distance of approximately $\sqrt{Dt} = 0.08$ cm during that interval, two orders of magnitude larger than the diameter of the focal region. The fraction which remain in the focal region should be on the order of $\frac{d^3}{(Dt)^{3/2}}$, or only $\sim 10^{-6}$. The number of dimers produced by a single pulse would thus have to be quite large for there to (a) be any dimers remaining in the focal region by the next pulse, and (b) a significant chance of a remaining dimer being in close enough proximity to 5 photons from the laser. For these reasons, we consider it unlikely that dimers are the source of the priming electrons.
Chapter 5

Experiments to Study Negative and Positive Charge Carriers

In this chapter I will describe three separate experiments which share many of the same features. Each was performed in the time of flight apparatus described in Chapter 3, with a focus on measuring the mobilities of positive and negative exotic charge carriers in liquid helium. Through these experiments I will show how these types of mobility measurements can serve as a probe into a wide range of interesting topics, including the origins of the exotic charge carriers, the nature of the wave function, and molecular cluster structures.
5.1 Pressure Dependence of the Exotic Ion Mobilities

5.1.1 Background

As mentioned in the Introduction, previous experiments have revealed 18 negatively charged ions with specific mobilities greater than that of the normal electron bubble [1, 3, 4, 21, 22, 23]. Though these objects have been observed by research groups working in four different laboratories, only two physical properties of the ions have been measured. The first is the mobility, which has been measured as a function of temperature. The mobility of the ions as a function of temperature has been found to vary as \( \mu \propto e^{\Delta/kT} \), consistent with the assumption that the mobility is limited by the rate of collisions with rotons. Thus, taking into account the cross section of the ions, the mobility should vary as \( \mu \propto e^{\Delta/kT} R^2 \), where \( R \) is the ion radius. By comparing the mobility of the different exotic ions with the mobility of the normal electron bubble, the radius of each ion can be estimated. The other measured property of the ions is the critical velocity at which they nucleate vortex lines. This critical velocity is found to increase with a decrease in the ion radius, and is in agreement with theoretical predictions. As previously mentioned, it was found that the fast ion, the fastest of all the observed charge carriers, does not produce vortex lines.

Though there have been many proposed origins for the exotic ions (impurities, helium anions, fractional electron bubbles) none have proven definitive. These explanations imply drastically different structures, which fit into two main categories, clusters and bubbles. Bubbles form when the charges are very mobile. In this case, the motion of the charge generates a large zero point pressure, which pushes the bulk liquid away creating a cavity. Clusters form when the charges are immobile. Because of the low mobility of the excess charge, little outward pressure is exerted on the neutral atoms in the bulk liquid. In this case the polarization force exerted by the
ion is dominant, drawing in and solidifying nearby atoms from the liquid.

To help understand the origin and structure of the different exotic ions it is important to measure some other property of the ions. We have attempted to do this by measurement of the mobility as a function of pressure. A change in the mobility with pressure will come from a change in the ion radius but there will also be a change in mobility from the change in the roton number density. If the change in the roton density can be allowed for, then the mobility can be used to give information about how the radius varies with pressure, and thus give insight into the structure of these objects. Here I present a summary of such an experiment designed to search for exotic ions and study how their geometry changes under external pressure.

Two early research groups, Keshishev et al. [52] and Meyer et al. [53, 54], performed experiments to measure the electron bubble and normal He$^+$ snowball mobilities under a wide range of pressure from saturated vapor pressure (SVP) up to solidification. More recent experiments by Brody have measured the mobility of both charge carriers in the greatest detail to date. Brody designed a time of flight experiment which utilized a 5.39 cm long drift field and a radioactive source [7]. The drift field was maintained at around 100 V/cm. The mobilities for both the normal electron bubble and positive helium snowball were measured along isotherms (1.375 K - 1.83 K) while the pressure was varied from SVP up to approximately 27.6 bar. The data of Brody has been included here in Fig. 5.1. The mobility in Fig. 5.1 is presented in terms of the friction coefficient $\Gamma = e/\mu$, where $e$ is the electric charge.

As previously discussed in Chapter 1 (Eqn. 1.1), the energy of the bubble consists of three terms,

$$E = \frac{\hbar^2}{8mR^2} + 4\pi\alpha R^2 + \frac{4}{3}\pi R^3 P.$$ 

The mobility of the ions as a function of temperature is well fit by $\mu \propto e^{\Delta/kT}/R^2$. The exponential factor accounts for the scattering of the ion with rotons. Neutron scatter-
Figure 5.1: Normal bubble (left) and helium snowball (right) mobility data of Brody [7].

Experiments show that the roton energy gap, $\Delta$, decreases roughly linearly with pressure [55]. So, as the density of the fluid increases, so will the density of rotons, producing more scattering and thus pushing the mobility to be smaller. The cross section term, meanwhile, decreases as the bubble compresses due to the pressure, but at a decreasing rate. As the surface tension and pressure terms of the energy push the bubble towards collapse, the zero point energy must compensate to maintain stability. However, as the pressure is increased the radius needs to decrease at a smaller and smaller rate for the zero point energy to compensate due to the $R^{-2}$ dependence of that term. These competing effects produce the minima visible in the left panel of Fig. 5.1. The shrinking of the electron bubble initially dominates up until approximately 2.8 bar, increasing the mobility, but it is then overpowered by the effect of increased roton scattering.

The clusters behave quite differently. The Atkins model posits that a stationary
excess charge will form a solid cluster of about 40 neutral helium atoms. This dense object has a radius of approximately 7 Å [15]. This is because the electric field polarizes the nearby neutral helium atoms. The polarized helium atoms become attracted to the core, and if this attractive force is strong enough at some radius the helium liquid will solidify there. Atkins showed that the pressure in the region of the cluster at a radius $r$ from the center can be approximated as

$$P(r) - P_o = \frac{N\alpha \epsilon_o e^2}{2V_o \epsilon_o^2 r^4},$$

(5.1)

and so the pressure experienced by the neutral atoms from the ionic core falls off as $P \propto 1/r^4$ [15]. Here $N$ is Avogadro’s number, $\alpha$ the molar polarizability, $V_o$ the volume per atom, and $\epsilon_o$ the vacuum permittivity. Inverting this equation and substituting $P_{solid}$, the helium solidification pressure near 1 K ($\sim 2.5$ MPa) for $P(r)$, we can solve for the cluster radius,

$$R^4_c = \frac{N\alpha \epsilon_o e^2}{2V_o \epsilon_o^2 (P_{solid} - P_o)}.$$  

(5.2)

From this we can consider how a cluster might behave under the application of external pressure. As the external pressure is increased, the radius $r$ at which $P(r) > P_{solid}$ grows. If $r$ becomes large enough such that another atom can join the solid core, the cluster will grow in size. Thus, for a cluster type object, we expect the radius to either remain constant or grow when pressure is applied.

These are, of course, only the extreme cases of charges which are free and highly mobile and those which are tightly bound and immobile. In reality the exact geometry depends on the wave function of the central impurity. For instance, an impurity ion with an excess electron which is only very loosely bound would, by the same argument as the normal electron bubble, push helium away and create a cavity. Bound electrons which orbit at intermediate radii might create hybrid objects, i.e. a small cavity with
a solid region just outside. An additional consideration is that bound electrons might migrate from atomic to molecular orbits as the solid core forms, further modifying the geometric configuration.

Unfortunately, there is no definitive model for the snowball structures. As such, we do not have a good estimate of the change in core radius that would be expected for the pressure range of this experiment. Regardless, the two structures are predicted to have markedly different behaviors under application of pressure. The bubbles should decrease in size, while the clusters remain the same or grow.

### 5.1.2 Experiment

The experiment was performed in the 6.15 cm long drift field cell described in Chapter 4. To reach pressures above the SVP, helium from an ultra high purity gas cylinder was forced through the cell fill lines. The pressure inside the cell was monitored by an analog gauge at room temperature outside the cryostat. To increase the pressure, the cylinder leak valve was further opened in very small increments. The pressure of the fluid inside the cell and the room temperature vapor in the fill lines come to equilibrium slowly. To account for this we waited at least 15 minutes, but often took as long as 60 minutes after making an adjustment to the pressure. This step ensured the pressure was not changing noticeably ($\pm 0.03$ bar) while a particular data point was being measured. The temperature was held constant at $T = 0.989$ K. In this experiment, the nylon spacer between S and G0 (described in 3.1.1, Fig. 3.2) was replaced by a nylon spacer of slightly different dimensions. The spacer used here is of thickness $1.5$ mm and inner diameter $2.1$ cm. The components of the experiment are otherwise identical to that described in Chapter 3.

This experiment was designed to allow measurement of the pressure dependence of any of the exotic ions. To make measurements under pressure it is, of course, necessary to work with the cell full of liquid and so there cannot be a plasma discharge.
Unfortunately, despite searching over a large range of voltage parameters, only the fast ion was visible under this condition. Varying the voltages on the grids did not make any of the other ions appear. This was not completely unexpected. The exotic ions have to date only been detected when a vapor discharge is present. The fast ion, on the other hand, has been detected in experiments in which $\alpha$-particles or electrons are emitted directly into the liquid [1, 25], as is the case here. The mobility of the fast ion as a function of pressure 0.989 K is shown on the right side of Fig. 5.2. We are limited to the pressure range below 5.5 bar because the cell was not designed to be used at higher pressures. The left side of Fig. 5.2 shows the mobility of the normal electron bubble at the same temperature and over the same pressure range.

Data for the normal electron bubbles and fast ion were recorded in two slightly different configurations. For both, a normal voltage configuration in which the radioactive Am$^{241}$ was held more negative than G0 was used. $V_{G2}$ was maintained at -400 V so that the drift field was 65 V/cm, while $V_{G1}$ was held at -390V. A gate pulse of amplitude -40V and width 1.0 ms was applied to G0 and G1 in all cases. The main difference lies in the voltage applied to G0. The fast ion was best resolved with $V_{G0} = -600$V, however in this configuration the signal from the normal electron bubble was not much greater than the noise. The electron bubble, however, was well resolved with $V_{G0} = -460$V. Both voltage parameter sets were used to measure the mobilities separately at each pressure. The applied voltage on the source varied between -1600 V and -2000 V. Both the normal electron bubble and fast ion signals became weaker with increasing pressure, and so the applied voltage on S was occasionally increased for better signal to noise.

Most noticeable is the overall similarity to the results of Brody, with a quadratic dependence from 0 - 5.5 bars for the normal electron bubble, and a linear dependence for the fast ion. The roton density is a property of the helium, and so it should not vary with ion species. Therefore, we can redefine the mobility $\mu$ in terms of a pressure
and temperature dependent drag coefficient $D(P, T)$ at temperature $T$ and pressure $P$ as

$$\mu_{\text{normal}}(P, T) = \frac{1}{D(P, T)R_{\text{normal}}^2(P)}. \quad (5.3)$$

Thus, we can write the fast ion radius in terms of the radius of the normal electron bubble and normal bubble and fast ion mobilities,

$$R_{\text{fast}}(P) = R_{\text{normal}}(P) \sqrt{\frac{\mu_{\text{normal}}(P, T)}{\mu_{\text{fast}}(P, T)}}. \quad (5.4)$$

Here we pause to mention an important detail. To estimate the radius as we are going to here, it must be assumed that the fast ion is a bubble type object. The magnitudes of the mobilities of the positive and negative charge carriers depend differently on the radius of the charge carrier. For instance, the mobility of the normal electron bubble is approximately 70% that of the He$^+$ snowball at zero pressure and near $T = 1$ K, yet the He$^+$ snowball cross section is only about 16% that of the normal bubble. This is presumably due to differences in how the rotons interact.
with the surface of the bubble as compared to the dense helium atoms that compose the cluster. It would be interesting to perform the same analysis assuming a cluster object, but we do not currently possess mobility data for the He\textsuperscript{+} snowball at these pressures and temperatures.

We use the method described in the previous section to calculate the normal electron bubble radius $R_{\text{normal}}$ at each pressure, using the measured value of the surface tension at $T = 1.0$ K of $\alpha = 0.347$ erg cm\textsuperscript{-2}\textsuperscript{[56]}. Figure 5.3 shows the calculated bubble radius for the pressure range over which the mobilities were measured in this experiment.

With this estimate of how the bubble radius varies with pressure in hand, we can calculate the fast ion radius using Eqn. 5.4. As shown in Fig. 5.4, the fast ion radius appears to vary only by approximately $5.6 \times 10^{-3}$ Å per bar increase in bulk pressure, despite the normal electron bubble decreasing by 18\% when the maximum 5.5 bars of pressure is applied. Thus, we find that, assuming the fast ion is a bubble like object, it has a radius of $R_{\text{fast}} = 7.5$ Å, and changes negligibly over the observed pressure range.
5.1.3 Discussion

There have been a number of proposed origins for the fast ion. These include the negative helium anion He\(^-\), impurity ions, and electron bubbles containing a fraction of the wave function.

An oft proposed explanation for the fast and exotic ions is impurity atoms which had been frozen on the cell walls. This is inconsistent with the available results in a number of ways, however. First, the exotic ions have been observed in a number of independent laboratories, in unique experiments, using different generation techniques. It is difficult to hypothesize a set 18 or more impurities with the right mobilities which would consistently remain on experimental parts of different materials constructed independently. Additionally, experimental advancements have only increased the number of observed exotic ions. If the ions were impurities, one might expect different subsets of the ions to show up from experiment to experiment. Second, while sputtering might dislodge frozen impurities from the walls when there is a plasma present, the fast ion has been generated with radioactive sources and sharp
tips submerged in the liquid. This indicates the fast ion is produced through interactions of energetic particles, either the alpha particles themselves or ejected electrons and helium ions, with the neutral helium atoms in the bulk liquid.

Shortly after the initial discovery of the fast ion by Doake and Gribbon [1], Ihase and Sanders suggested that the fast ion might consist of a negative helium anion $\text{He}^-$ [21] in the $^4P_J$ state. This excited state is formed through the process $\text{He}^* + e^- \rightarrow \text{He}^-$. There remains a major issue with this explanation. The excited ion state is short lived in all momentum configurations. The $J = 5/2$ state has a lifetime of 345 $\mu$s and the $J = 3/2, 1/2$ states of 11 $\mu$s [57]. The drift time in the cell for the fast ions is at least 4 ms, and so almost all should decay before they reach the collector. There have however been no detailed measurements of the anion lifetime in the liquid, and the ultimate configuration might inhibit the decay process $\text{He}^- \rightarrow e^- + \text{He}$. For instance, one potential mechanism involves the 1 eV energy barrier for an electron to enter the bulk liquid. The ejected electron might be reflected at the interface between the anion and the bulk liquid, and thus forced to remain in the excited bound state. This seems unlikely, however, due to the ejection energy (19.8 eV) being large compared to the 1 eV barrier.

We can also consider the implications of the electron fission model [8]. A bubble containing only a fraction $F$ of the wave function will have total energy,

$$E = \frac{F h^2}{8 m R} + 4 \pi \alpha R^2 + \frac{4}{3} \pi R^3 P$$

An electron bubble with a 7.5 Å radius corresponds to a fraction of the wave function $F = 0.022$. Once again, we minimize the energy to estimate the change in radius with pressure and find that the bubble radius should decrease by 10% over the measured pressure range. This is inconsistent with our results.

Finally, it is interesting to take a more phenomenological approach. Here we discuss a procedure similar to that used by Wei et al. [8]. Let us suppose that there
is a negative ion in the liquid; we don’t ask whether it is an impurity or some ion of helium that has not been discovered. The ion must have properties such that when immersed in the liquid it forms a structure with a radius of about 7 Å. How much does the radius of such an object change when a pressure is applied? Is it a bubble or is a solid cluster of helium atoms?

Let’s first consider the bubbles, and construct a simple potential which consists of a finite well and takes into account the repulsive force at the ion nucleus and the 1 eV helium barrier at the liquid interface. We have a potential $V(r)$ defined as,

$$V(r) = \begin{cases} 
-V_o, & r < a \\
0, & a < r < R \\
\infty, & R < r
\end{cases}$$

(5.6)

where $a$ is the width of the potential well, $V_o$ is its depth, and $R$ is the ion radius.

First, let’s consider the case of a pure bubble state. For $a = 2.0$ Å, we find that a bubble of $R = 7.0$ Å requires a well depth $V_o = -4.3$ eV. The electron affinity of this hypothetical ion is $\phi = 0.63$ eV. For each set of well parameters, we can calculate the bubble energy $E_{el}$ as a function of bubble radius. By replacing the zero point energy term of Eqn. [1.1] with $E_{el}$, we can then adjust the external pressure and determine how the bubble radius is expected to change. Doing this, we find that the bubble radius should shrink by approximately 6%, from 7 Å to 6.6 Å. As an additional consideration, we performed this analysis while including the polarization energy term, $-\left(\epsilon - 1\right)e^2/2\epsilon R$, where $\epsilon$ is the dielectric constant. This lowered $E_{el}$ by around 20% but had a negligible impact on the change in the ion radius.

Second, we can consider what physical properties an Atkins type cluster with the mobility of the fast ion would require, and what those properties imply about the
pressure dependent behavior of the cluster. In this section we estimated the fast ion radius (assuming it is a bubble) from the normal electron bubble mobility. We can perform a more limited but similar calculation, this time assuming the fast ion is a cluster like object, and use measurements of the He$^+$ mobility to estimate the cluster size. At $P = 0$ bar and $T = 0.989$ K, we have $R^{\text{cluster}}_{\text{fast}} = 3.7$ Å. Using Eqn. 5.2 for the cluster radius, we find that for a cluster of this size, the radius would grow to 3.88 Å under the application of a pressure of 5.5 bar.

In conclusion, we find that the fast ion mobility’s dependence on pressure is consistent with it either being a dense cluster or hybrid type object, rather than a bubble. This is an especially interesting detail when considered in the context of previous fast ion observations, namely how it is generated. Though the fast ion has been observed in experiments in which the exotic ions were present, it is the only negative ion of discrete mobility less than the normal bubble which has been produced alone, or with the source submerged in the liquid. These facts imply an origin related to interactions in the liquid with constituents that are present in all experiments (i.e., neutral helium, ionic helium, helium molecules, free electrons).
5.2 Study of the Type 2 Background

Below follows the details of an experiment designed to examine the previously observed “#2” background, a distribution of charge carriers with continuous mobility present in the exotic ion measurements of Wei et al. [8]. In those experiments, and these detailed here, electrons are transmitted from helium vapor into liquid under the influence of a DC electric field. Those researchers proposed that the #2 background is the product of electrons with energy near that of the barrier for entry into the liquid being only partially transmitted across the boundary. The fraction of the wave function which is transmitted forms a bubble of size smaller than that of the normal electron bubble, and that part of the wave function thus becomes trapped. Because of uncertainty in properly measuring the “leakage” signal caused by the imperfect nature of the Frisch grid, Wei et al. did not insist on this explanation. In this section I detail new measurements of this background, present a computational model for the leakage signal, and show that the expected electron energy distribution in the plasma gives rise to a signal consistent with the #2 background.

These results have been submitted for publication [58], and some portions of the text and some figures have been excerpted directly here.

5.2.1 Background

The measurement process in quantum mechanics remains of great interest. According to the Copenhagen interpretation of quantum mechanics when a quantity \( f \) is measured, the result of the measurement must be one of the eigenvalues \( \{ f_n \} \) of the operator \( \hat{f} \). In addition, if the result of the measurement is \( \{ f_n \} \) the act of making the measurement results in the wave function instantly changing to the eigenfunction \( \phi_n \) corresponding to the eigenvalue \( f_n \). This leads to a number of unresolved issues. Putnam argues that quantum mechanics “treats the universe as consisting of two kinds
of objects – classical objects not subject to uncertainties and micro objects – with
the former measuring the latter” [59]. He points out that this is, of course, inconsis-
tent with the idea that quantum theory describes all objects. These questions were
discussed many years ago, for example in the papers by Putnam [60], by Margenau
and Wigner [61,62,63], and by Cooper and Van Vechten [64,65,66]. Another obvious
question concerns relativity [67]. Suppose that at time $t$ a measurement is made at
a point $\vec{r}$ to look for a particle, and the particle is found to be there. The wave
function is supposed to immediately collapse and become zero everywhere except at
$\vec{r}$. This may require that news of the measurement travels faster than the speed of
light.

We can also investigate another aspect of the measurement problem. The concept
of the experiment is shown in its simplest possible form in Fig. 5.5. Suppose that we
trap a part of the wave function of an electron in a box. The box could be a hollow
cylinder of radius $R$ closed at its lower end by a fixed plate. At the top of the cylinder
is a sliding piston of mass $M$. The equilibrium position of the piston is at the height
$L$ at which there is a balance between the gravitational force of magnitude $Mg$ and
the upward force on the piston due to the zero point energy of the trapped electron.
If the electron is in its ground state and the integral of $|\psi|^2$ over the part of the wave
function that is trapped in the cylinder is $F$ then

$$L = \left( \frac{F \hbar^2 \pi^2}{m M g} \right)^{1/3} \quad (5.7)$$

where $m$ is the electron mass. This assumes that the energy of the part of the wave
function not in the cylinder is not affected by the position of the piston. In an
apparatus like this the electron will be constantly interacting with the material on
the wall of the cylinder. Does this result in a measurement that determines that the
electron is either in the box or not? (A more detailed discussion of this question is
provided in [68]) If there is no measurement the position will be as given by Eq. (5.7).
Figure 5.5: Concept of the experiment. An electron is trapped inside a cylinder with a movable piston.

If there is a measurement the final position of the piston is either at $L = 0$ or at $L = (\hbar^2 \pi^2 / mMg)^{1/3}$.

How can one perform an experiment to trap a part of the wave function in a box? There has to be a means for directing the wave function to some region, and then changing the potential acting on the particle sufficiently rapidly so that a part of the wave function becomes trapped. It might be possible to do this with some type of semiconductor structure using voltage-controlled gates. The experiment also has to be able to determine at a later time whether the wave function remains trapped, i.e., the apparatus has to include some element that plays the role of the piston in Fig. 5.5. In the experiments reported here we use a simple method in which the trapping takes place automatically and there is a straightforward way to tell whether the wave function is still trapped at a later time.

In the experiments electrons are produced in a plasma discharge in the helium
vapor above a volume of liquid helium. In order to enter the liquid from the plasma an electron has to overcome a barrier of height $V_o$ which is approximately 1 eV [12 13]. Thus, the minimum energy an electron can have while propagating through uniform liquid is 1 eV. However, once in the liquid the electron can reach a lower energy state by forcing open a spherical cavity and becoming trapped in it. To a good approximation the wave function of the trapped electron is

$$\psi = \sqrt{\frac{1}{2\pi R}} \frac{\sin(\pi r/R)}{r}$$

where $R$ is the cavity radius. The total energy of the bubble state is then

$$E_{\text{bubble}} = \frac{\hbar^2}{8mR^2} + 4\pi R^2 \alpha$$

where $m$ is the mass of the electron, and $\alpha$ is the surface tension of the liquid. The first term is the zero-point energy of an electron in a spherical cavity; the penetration of the wave function into the liquid is small and has been neglected. The second term is the energy of the bubble surface. The total energy is a minimum when

$$R = \left(\frac{\hbar^2}{32\pi m\alpha}\right)^{1/4}.$$ 

The surface tension of liquid helium at $T = 0$ K is 0.375 erg cm$^{-2}$ and this gives the radius for minimum energy to be 19 Å. From this we obtain the energy of the trapped electron plus the energy of the bubble wall as

$$E_{\text{bubble}} = \hbar \sqrt{\frac{2\pi \alpha}{m}} \approx 0.21\text{eV}. \quad (5.10)$$

This is much less than $V_o$.

The trapping process is shown schematically in Fig. 5.6. An electron from the plasma is incident on the free surface of the liquid helium. The electrons in the plasma have a continuous distribution of energy, and if the energy of the electron is greater than $V_o$, a part of the wave function of the electron will pass into the liquid and a part will be reflected (“reflection above the barrier”). Let the integral of $|\psi|^2$ over
Figure 5.6: Schematic diagram of the trapping process in liquid helium. The solid line shows the wave function of the electron and the dashed line is the density of the liquid helium. The bulk liquid occupies the right hand part of the figure. The potential acting on the electron is approximately proportional to the helium density. In (a) an electron approaches the free surface of the liquid with an energy above the barrier. A part of the wave function is reflected (b) and a part enters the bulk liquid. A bubble is formed in the liquid (c), and grows to its full size (d).
the region containing the part of the wave function that goes into the liquid be $F$. This part of $\psi$ will lose kinetic energy by interactions with helium atoms and after traveling a distance of the order of 100 Å will have slowed down sufficiently to form a bubble. Because of the high barrier provided by the helium, the parts of the wave function inside and outside are disconnected. The part inside will form a bubble of radius $R_\%$ and have a wave function which is

$$\psi = \sqrt{\frac{F}{2\pi R_\%}} \sin(\pi r/R_\%) \frac{r}{r}$$  \hspace{1cm} (5.11)$$

Since the wave function penetrates a negligible distance into the bubble wall, the potential energy is zero and the electron energy is

$$E_\% = \int \frac{\hbar^2}{2m} |\nabla \psi|^2 dV = \frac{F\hbar^2}{8mR^2_\%}$$  \hspace{1cm} (5.12)$$

and so the total energy is

$$E_{\text{bubble}}^\% = \frac{F\hbar^2}{8mR^2_\%} + 4\pi R^2_\% \alpha$$  \hspace{1cm} (5.13)$$

Then the lowest energy state is with a bubble radius of

$$R_\% = RF^{1/4}.$$  \hspace{1cm} (5.14)$$

Since, as discussed below, the electrons in the plasma have a continuous distribution of energy, electron bubbles with a continuous size distribution should be produced. To estimate the distribution of bubble size it is necessary to consider the energy spectrum of the electrons in the plasma and how the fraction of the wave function that is transmitted varies with the electron energy.

Note that the above discussion assumes that no measurement is made by the helium even though the electron wave function in the bubble is in contact with the
helium atoms on the wall of the bubble. If the helium did make a measurement and found an electron, a bubble of normal size (radius $R$) would result. If no electron were found by the measurement, then there would be no bubble.

Mobility measurements provide a straightforward way to detect the existence of electron bubbles of different size. In superfluid helium at temperatures in the vicinity of 1 K, a moving bubble experiences a drag force due to the scattering of thermally-excited rotons. To a reasonable approximation the drag force is proportional to the roton number density and to the square of the bubble radius $[14]$. Thus a measurement of the mobility $\mu$ can be used to find the bubble radius and look for a continuous distribution of bubble size resulting from the mechanism described above.

### 5.2.2 Summary of Previous Results

Here I present a brief analysis of the key components of the negative charge carrier spectrum. A similar breakdown was previously provided by Wei et al. $[8]$. This overview is a summary of experiments performed by four different sets of researchers, Doake and Gibbon, Ihas and Sanders, Eden and McClintock, and Wei et al. These experiments were previously discussed in the Chapter 1, but here we will focus more on the main characteristics of the discharges and output signals. These experiments all shared a basic structure, namely an electron source, a stable drift field for the charge carriers to travel through, and a charge collector for making time resolved measurements of their arrival. Within this template, however, these experiments have shown that a number of critical parameters greatly affect what carriers are produced and with what strength. These can (largely) be broken down into three groups.

- **Production of Ionization**

  Doake and Gibbon used an $\alpha$-source which was submerged in the liquid $[1]$. In this case, the $\alpha$-particles only travel a short distance on the order of 100 $\mu$m.
Ihas and Sanders performed experiments using either sharp tungsten tips or a beta source placed above the liquid interface \[2, 21, 22\]. The sharp tips are held at a voltage such that they produce electrons by field emission. Tungsten tips above the liquid were also utilized in the experiments of Eden and McClintock \[3, 23, 24\]. Finally, sharp tips consisting of carbon nanotubes were used in a series of experiments by Wei et al., placed both above and under the liquid interface \[4, 5, 8, 26\].

- **Cell Geometry**

  The design of the source itself, the design of the electrodes in proximity to the source, the spacing between these elements, and the materials chosen will all affect the discharge characteristics. The position of the gas-liquid interface, for example, had a strong influence on which ions were produced in the experiments of Ihas and Sanders \[21\] and Eden and McClintock \[23\].

- **Discharge Parameters**

  The discharge has, at minimum, two independent voltages for the source and another electrode. The cell geometry can give rise to quite complicated electric field behaviors. In our experiments there exists a critical voltage for plasma production. This point is characterized by a sudden voltage "drop" for the source, as well as visible light emanating from the discharge region. There are additional critical points at higher voltages, which typically result in increased heat generation, changes in the visible light spectrum, and changes in ion production.

  The charge carrier spectrum was broken down into four components previously by Wei et al. \[8\]. The largest, most consistent feature is the so called normal ion (N) peak. This peak originates from electrons which enter the bulk liquid and form \(\sim 19\) \(\text{Å}\) cavities, the so called normal electron bubbles. While the overall normal ion signal varies, they are generally easy to observe under a wide variety of discharge conditions.
Figure 5.7: Solid curve shows the current arriving at the collector as a function of time. Measurements are by Wei et al. [8], temperature is 0.991 K and the drift field is 82.1 V/cm. Dashed curve shows the result of removing each of the peaks from the data. The large peak labeled NB arises from normal electron bubbles.

Fig. 5.7 shows data replotted from the experiments of Wei et al. as a solid line. This particular dataset was produced using tungsten tips at a temperature of 0.991 K. The drift field portion of the cell was 6.15 cm long, identical to that used here. The drift field itself was 82.1 V cm\(^{-1}\). In Fig. 5.7 the peak produced by the normal bubbles is labeled “NB”. The normal bubble mobility measured by Wei et al. was in good agreement with previous measurements [13].

The second component is the exotic ions. The exotic ions are characterized by discrete peaks which arrive at times earlier than the normal bubbles. These are objects which are almost always much weaker than the normal ion signal, but with larger mobilities. The “fast ion” was the first of the exotic ions to be discovered, in an experiment performed by Doake and Gribbon [1] using an alpha source. It was the only exotic ion observed in their experiment. The fast ion has the highest mobility of any negative ion observed to date. Subsequent experiments by Ihhas and Sanders [21], utilizing sharp tungsten tips as the electron source, revealed 13 more
exotic ions in addition to the fast ion. Most recently, Wei et al. [8] used carbon nanotube tips to observe 18 exotic ions, 13 of which were those previously observed. The exotic ions are the component most sensitive to the discharge conditions discussed above. Although the mobilities of the exotic ions are entirely determined by the drift field conditions, their amplitudes vary greatly with the discharge conditions. In fact, many discharge conditions produce no observable exotic ions. There is, thus far, no definitive explanation for the exotic ions. Proposed explanations have include impurities, helium ions, and fission of the electron wave function. These are discussed in some depth in Wei et al. [8].

The third component is referred to as the #1 background. The geometry of the signal peaks is determined by a complex interaction of the various electrical fields in the discharge region, any space charge accumulating near the liquid vapor interface, the characteristics of the gate pulse, and the interaction of the ions with the Frisch grid and collector. As such, there is no complete model for their shape. However, by fitting the peaks with a simplified model and subtracting the fits from the signal, Wei et al. showed there exists a background which varies continuously with mobility [8]. This is shown as the dashed line in Fig. 5.8. The #1 background is particularly interesting in that it features a sharp beginning at approximately half the arrival time of the normal bubble, and that this mobility varies with temperature in the same way as the exotic ions, $\mu \propto e^{\Delta/RT}$. This implies the existence of a set of ions with continuous mobility, and thus continuous size. Wei et al. found that this #1 background is always present when exotic ions are observed, but did not observe the #1 background when exotic ions were absent under any conditions.

The fourth and final component is referred to as the #2 background. Wei et al. found that when both the exotic ions and #1 background were absent, there was yet another continuous background signal. By adjusting the applied voltage to modify the field near the source slightly, the #1 background could be turned on and off.
Figure 5.8: Current arriving at the collector as a function of time from Wei et al. [8]. The temperature is 1.025 K and the curves are labeled by the drift field. The signal due to normal electron bubbles is labeled as NB. The dashed curves show data taken when background of type #1 is present, and the solid curves are taken when the voltages on the plasma have been changed by a small amount to make background #1 disappear.

Data recorded at a temperature of 1.025 K by Wei et al. showing this phenomenon is plotted in Fig. 5.8. The solid lines, where no #1 background is present, shows a signal with continuous mobility up to $\mu = \mu_{NB}$. The signal also increases monotonically in this direction. Like the #1 background, this observation implies a set of ions with a continuous size distribution. In the experiments performed thus far by Wei et al., and those discussed in the next section, the #2 background has always been observed when a) there is no #1 background, and b) there is a normal bubble signal. This indicates two important things. One, the signal likely originates from the interaction of the electrons with the surface of the liquid. Two, the #2 background is likely always present so long as electrons are entering the liquid, and is simply being masked by the exotic ion and #1 background signals in other cases.
Figure 5.9: Current arriving at the collector as a function of time. The temperature is 1.026 K and the drift field is 65.0 V/cm. The curves are labeled by the difference in the voltage applied between grids G0 and G1 in volts. The curves are offset for clarity. Data before 5 ms are not included because in this time range there is a large contribution to the collector signal coming from electrical pick up from the gate pulse applied to G1.

5.2.3 Experimental Results

The experimental observations of the #2 background detailed in this experiment are plotted in Fig. 5.9. The data have been plotted on an expanded scale so that the #2 background is clearly visible. All were recorded in the reverse voltage configuration, with no external voltage applied to S. By applying a large negative voltage to G0, a glow discharge was produced in the S-G0 region. G2 was held at constant voltage such that the drift field was 65 V/cm. All observations were made at a temperature of 1.026 K. The gate pulse applied was always -40 V in amplitude. The duration of the gate pulse was adjusted so as to maintain a similar normal electron bubble current as the voltage parameters were changed. The liquid to vapor interface was maintained at approximately 0.5 mm above G1. The field between G0 and G1 was varied by increasing the voltage on G0. This field, in part, determines the energy with which electrons impact the surface.
5.2.3.1 Background of the #2 Background

This fission model, in which the electron wave function can split and form different sized bubbles, was originally proposed as an explanation for the discrete exotic ions [69]. Wei et al. discussed the #2 background extensively in a previous paper [8], suggesting that the fission model, applied to the interaction between electrons in the plasma and the 1 eV helium interface potential barrier, should result in partial transmission of the wave function and thus a continuous distribution of bubbles up to the mobility of the NB. Although the fission model appeared to give a good fit to data, the authors decided at the time that the origin of the #2 background was unclear due to uncertainty in understanding the impact of the Frisch grid. As mentioned in Chapter 3, if there were no Frisch grid the ions would induce an image charge on the collector. A negative ion will induce a positive image charge, and so a positive charge will have to travel from ground and through the 2.5 MΩ resistor. This generates a negative voltage across the resistor, which is then picked up by the preamplifier. Because the Frisch grid is constructed from a wire mesh, there must be some electric field lines which leak through the gaps into the F-C region. The amount of field lines leaking through will presumably increase with decreasing distance from the Frisch grid. Thus, there is a reasonable concern that the time dependence of the leakage signal will resemble that of the #2 background. The normal bubble signal is much larger than the #2 background; The #2 background signals visible in Fig. 3.9, for example, are approximately 50 times smaller than the associated signal from the normal bubbles. Thus, it is critical to understand the implications of even a very small amount of field lines leaking through the Frisch grid. To investigate the possibility that leakage through the Frisch grid is the origin of the #2 background it is necessary to know how many field lines pass through the grid to the collector. This calculation was not attempted at the time because the geometry of the grid was uncertain; the only information available was that the repeat distance of the grid was 70 wires per
inch. Neither the thickness of the grid nor the shape of the wires was known.

In Chapter 3, I analyzed an idealized version of the process by which a finite slab of ions moves through the drift field and produces a signal on the collector. Now we first continue to treat the Frisch grid as a perfect electrical shield, while considering how those as well as any other effects determine the shape of a pulse from a slab of ions. These effects include:

1. the duration and magnitude of the voltage pulse applied to G1 in order to generate the ion pulse;
2. the velocity of the ions when moving under the influence of the drift field;
3. the inhomogeneity of the drift field which gives a "tail" after the peak of the detected pulse. Note that field inhomogeneity cannot give a tail before the peak.
4. the electric field coming from the space charge produced by the normal electron bubbles;
5. the passage of the ions through the Frisch grid and into the space between this grid and the collector.

The net result of these effects is a normal bubble full width at half maximum of approximately 3 ms, as seen in Fig. 5.9. Effects (1-4) affect the shape of the pulse before it reaches the Frisch grid. Effect (5), however, regards how the interaction of the collector structure and the ion slab affects the pulse shape. As a consequence of (5), the width of the leakage signal produced by a slab of ions as it moves down the drift field will be much larger than the width of the pulse that slab generates when it moves between the Frisch grid and collector. As a demonstration of this, consider the effect of a single slab of charge -Q, of infinitesimal thickness, which begins at G2 and then travels down the cell under the influence of a drift field $E_d$. We assume this thin disc of charge is centered on the axis of the drift field. This is shown schematically
in Fig. 5.10. There will be a current produced from the leakage of field lines over the entire path to the Frisch grid, and thus the width of the signal is the time of flight, \( t_d = \frac{z_d}{\mu E} \). Here \( z_d = 6.15 \) cm, the length of the drift field.

Now let’s consider what happens as the charge passes through the Frisch grid in the idealized, no leakage scenario. When there is complete shielding there will be no charge induced on the collector until the ions pass through the grid \[70\]. As discussed in Chapter 3, the induced current on the collector from a charge \(-Q\) in the F-C region is proportional to the amount of charge and the ion velocity. Thus, the signal produced over the entire path is:

\[
I(t) = \begin{cases} 
0, & t \lt t_{NB} - \frac{z_{FC}}{v_d} \\
= -Q \frac{v_{FC}}{z_{FC}}, & t_{NB} - \frac{z_{FC}}{v_{FC}} < t < t_{NB} \\
0, & t > t_{NB}
\end{cases}
\]

(5.15)

where \( z \) is the ion position with respect to the collector, \( z_{FC} \) is the F-C separation distance, and \( t_{NB} \) is the arrival time of the normal bubble. Here we are neglecting the small loss of ions across the Frisch grid mentioned previously in \[70\]. We are also neglecting the previously discussed small, \( \sim 170 \mu s \) rise time associated with the capacitance of the collection circuit. For the data shown in Fig. 5.9, \( z_{FC}/v_{FC} = 0.58 \) ms. This is much smaller than the observed 3 ms width of the normal bubble pulses. From this result we can surmise that the “real” pulse width, that of the normal bubbles as they travel through the drift field, is significantly less than that of the observed pulse (3 ms). This width is small compared to the time range over which the background #2 is evident (see Fig. 5.9), and so for the purposes of calculating the effect of leakage we can as a first approximation take the ion pulse as a slab of zero thickness. We mention some limitations of this approximation later in the discussion.
Figure 5.10: Schematic diagram showing a slab of charge approaching the Frisch grid and the collector. The dashed lines show field lines from the slab of ions.

Consider now the penetration of the field through the grid. We consider a thin slab of ions at distance from the collector and with a uniform ion density per unit area within a radial distance of 0.4 cm from the vertical cell axis, i.e., forming a slab with the same diameter as the grids G1 and G2. The total charge on the slab is taken to be equal to the integral of the detected current over the time range close to the arrival time of the normal electron bubbles. The problem then amounts to finding the number of field lines from these ions that pass through the Frisch grid and reach the collector. The detected current due to the leakage through the grid is then

$$I(t) = \frac{1}{4\pi} \frac{d}{dt} \int E dA.$$  \hspace{1cm} (5.16)

where the integral of the field $E$ is over the surface area of the detector. The distance of the slab from the collector at time $t$ is

$$z = v(t_{NB} - t)$$  \hspace{1cm} (5.17)
Figure 5.11: Scanning electron microscope images of the Frisch grid. Part (a) shows one cell of the grid, and (b) is an image taken with higher magnification showing a cross-section of one of the wires.

In order to obtain detailed information about the geometry of the grid structure, scanning electron microscope images of a section of the grid were made. Two examples of these images are shown in Fig. 5.11. For numerical calculations a slightly simplified description of the geometry was used. The grid was assumed to have a repeat distance of 326 µm in both the x and the y directions. The width and thickness of the grid wires were taken to be 38 µm and 13 µm, respectively. The rounding of the top corners of the grid was approximated by an arc of radius 13 µm. The bottom surface of the wires was taken to be flat, i.e., the small ridge seen in Fig. 5.11b was ignored. The geometry of the collector and the other parts of the cell, such as the homogenizer disks, can be measured in a straightforward way.

To calculate the field on the surface of the collector for a given position of the slab of ions we have used a Monte Carlo method. This particular approach is well-suited for the solution of electrostatics problems in which some of the elements have a much
smaller length scale than others; this is the case here.

To determine the effect of leakage it is necessary to find how many field lines from a charge $Q$ situated at a position $\vec{r}$ will reach the collector plate. To reach the collector these lines have to pass through the Frisch grid. This problem could possibly be addressed using standard methods based on finite element techniques. However this approach is made difficult because of the wide range of length scales that are involved. As shown in Fig. 5.11 the geometry of the field lines is influenced by the presence of conducting electrodes of dimensions of the order of centimeters (such as the homogenizer disks), and also by structures with features as small as $10 \, \mu\text{m}$ (the wires making up the grid). This wide range of size requires a complicated mesh.

The Monte Carlo method is as follows [71, 72]. Consider a charge located at some particular position in the cell. A “walker” starts at the position of the charge. The walker then undergoes a random walk within the volume occupied by the conducting electrodes. The random walk ends when the walker reaches an electrode. This procedure is then repeated for $N_{\text{walkers}}$ walkers, and the number $N_{\text{collector}}$ of these walkers that reach the collector, rather than some other electrode, is determined. The image charge on the collector is then given by

$$Q_{\text{image}} = -Q \frac{N_{\text{collector}}}{N_{\text{walkers}}} \quad (5.18)$$

This result by itself would not provide a very useful algorithm because, at first sight it would appear that to give an accurate result the steps of the random walk would have to be smaller than the smallest linear dimensions of any of the structures. If this were chosen to be $5 \, \mu\text{m}$ then for a walker to migrate a distance of $1 \, \text{cm}$ would require of the order of $10^7$ steps. However, it can be shown [71, 72] that the result of Eq. 5.18 still holds even if the step distance is varied during the random walk. The only requirement is that at each stage of the walk the step distance must be less than the distance $\xi$ from the current position of the walker to the nearest electrode. Thus
to make the required number of steps a minimum, in each step the walker is moved to a point randomly chosen on the surface of a sphere of radius $\xi$ centered on the previous position. In our program the random walk is terminated when the walker reaches a position which is less than 0.1 $\mu$m from an electrode. The justification for terminating the random walk slightly outside an electrode is given in section 7.2 of ref. [71]. Notice that the detailed path of the walker is not of interest.

The result for the ratio of the charge induced on the collector to the total charge on the slab is shown in Fig. 5.12. To obtain the results shown in Fig. 5.13 we used $2 \times 10^6$ walkers with initial positions distributed uniformly over the area of the slab of ions as shown in Fig. 5.11. This calculation was then repeated for a series of positions of the slab giving the results shown in Fig. 5.13. The leakage increases rapidly as the distance of the charge from the Frisch grid decreases, and the simulation results are well fit by the expression.

$$Q_{\text{leakage}} = -Qa \exp\left(-\frac{z}{b}\right)$$

where $Q_{\text{leakage}}$ is the charge induced on the collector by field leakage, $Q$ is the charge of the slab of ions, $a = 0.0375$, and $b = 0.355$ cm. Hence, the current due to leakage is

$$I_{\text{leakage}} = -\frac{dQ_{\text{leakage}}}{dt} = \frac{Qav}{b} \exp\left[-v\left(t_{NB} - t\right)/b\right]$$

where, as discussed above, $Q$ is equal to the integral of the detected current over the time range covering the arrival time of the normal electron bubbles, and $v$ is the velocity of the normal electron bubbles. The value of $Q$ is different for each data trace. For the traces taken when the voltage difference $\Delta V_{01}$ is between 20 and 80 V the value of $Q$ is in the range between -0.331 and -0.397 pC. At higher voltages the charge $Q$ begins to decrease rapidly; for $\Delta V_{01} = 90$ V the charge is -0.304 pC and
Figure 5.12: Plot of the ratio of the charge induced on the collector to the total charge of an approaching ion pulse as a function of the distance $z$. The diamonds are the results of the Monte Carlo simulations and the solid line is a fit to these results.

for $\Delta V_{01} = 100$ V it is -0.216 pC, and so the leakage is significantly smaller. From Fig. 5.9 we see that the peak in the normal electron bubble signal is at $t_{NB} = 32$ ms. In Fig. 5.13 we show the measured signal as the solid curve and the calculated leakage current as the dashed curve. It is clear that this result for $I_{\text{leakage}}$ is much too small to make a significant contribution to the signal over the time range in which the background #2 is seen. At a time of 25 ms, for example, the data for $\Delta V_{01} = 20$ V has a value of -1.32 pA, whereas the value from Eq. 5.20 is -0.16 pA.

5.2.3.2 Modeling the Plasma

We now compare these results with the signal expected from the mechanism described in section 5.2.1, i.e., from electrons which have a wave function which is only partially transmitted from the vapor into the liquid and hence form smaller bubbles in the liquid. The number of electrons in the energy range $\epsilon$ to $\epsilon + d\epsilon$ the plasma is given by
Figure 5.13: Plots of the measured type #2 background (solid curve) and the leakage through the Frisch grid (dashed curve). The plots are labeled by the voltage difference $\Delta V_{01}$ between grids G0 and G1.

\[ n(\epsilon)d\epsilon = \frac{2n_{el}}{\Gamma(3/4)} \frac{\epsilon^{1/2}}{\epsilon_o^{3/2}} \exp\left(-\frac{\epsilon^2}{\epsilon_o^2}\right)d\epsilon \]  \hspace{1cm} (5.21)

where $n_{el}$ is the total number of electrons per unit volume and

\[ \epsilon_o = \Lambda e E \sqrt{\frac{M}{3m}} \]  \hspace{1cm} (5.22)

where $\Lambda$ is the electron mean free path, $E$ is the applied field, and $M$ is the helium mass \([73]\). The mean free path is

\[ \Lambda = \frac{1}{n_{He}\sigma_{el-He}} \]  \hspace{1cm} (5.23)

where $n_{He}$ is the number density of helium atoms and $\sigma_{el-He}$ is the cross-section for electron-helium scattering. This cross-section is $\sim 5 \times 10^{-16}$ cm$^{-2}$ \([74]\). The data in Fig. 5.9 were taken at 1.026 K where the helium number density is $1.40 \times 10^{18}$ cm$^{-3}$ and the mean free path is therefore $1.43 \times 10^{-3}$ cm. For Eq. 5.21 to be valid
the electron energies must be large compared to \( kT \); this is the situation in our experiment. In addition it is assumed that the electrons lose energy primarily by quasi-elastic collisions with helium atoms rather than by exciting atoms out of the ground state.

The number of electrons in a volume element \( dk_xdk_ydk_z \) of \( \hat{k} \) space is then

\[
\frac{h^3n_{el}}{\sqrt{32\pi\Gamma(3/4)m^{3/2}e_o^{3/2}}} \exp(-h^4k^4/4m^2e_o^2)dk_xdk_ydk_z.
\] (5.24)

The rate at which electrons are incident on the vapor-liquid interface is

\[
\int_{-\infty}^{\infty} dk_x \int_{-\infty}^{\infty} dk_y \int_{0}^{\infty} dk_z \frac{h^3n_{el}}{\sqrt{32\pi\Gamma(3/4)m^{3/2}e_o^{3/2}}} \exp(-h^4k^4/4m^2e_o^2)k_zA
\] (5.25)

where \( A \) is the area of the surface. The density of the liquid helium in the immediate vicinity of the interface has been measured and calculated \([75]\) and can be reasonably well described by \( \rho(z) = \rho_o/[1 + \exp(-z/a)] \) where \( \rho_o \) is the mass density in the bulk liquid and \( a = 1.59 \text{ Å} \) \([4]\). We assume (see also later discussion) that the potential \( V(z) \) experienced by an electron incident on the interface is proportional to the density and so

\[
V(z) = \frac{V_o}{1 + \exp(-z/a)}
\] (5.26)

where \( V_o \) is 1 eV. An electron incident on the interface with a normal momentum \( k_z \) less than \( \sqrt{2mV_o/h^2} \) will be completely reflected. If \( k_z > \sqrt{2mV_o/h^2} \) then the fraction \( F \) of \( \psi^2 \) that is transmitted is

\[
F = \frac{\sinh[2\pi k_z a] \sinh[2\pi k'_z a]}{\sinh^2[2\pi(k_z + k'_z) a]}
\] (5.27)

where \( k'_z = \sqrt{k_z^2 - 2mV_o/h^2} \) \([76]\).
Using the last three equations we can make a histogram for how the number of electrons with a particular value of $F$ varies with $F$. This can then be used to find the number of electrons with a transit time through the liquid in the drift cell in a given range, i.e., the current as a function of time. The result for this current is proportional to the number density $n_{el}$ of electrons in the plasma which is not known. To fix the value of $n_{el}$ we can use the requirement that the integral of the calculated current over the full range of transit times must be equal to the experimental value. This leaves $\epsilon_o$ as the only adjustable parameter. If the field $E_{vap}$ in the vapor just above the liquid surface were known then $\epsilon_o$ could be obtained from Eq. 5.22. However, although the voltage difference $\Delta V_{01}$ between G0 and G1 is known, it is not possible to make a reliable estimate of $E_{vap}$ because of the following effects:

(a) Because of the energy barrier that electrons have to overcome in order to enter the liquid, there will be a layer of charge at the liquid-vapor interface. The amount of charge will depend on the value of $\Delta V_{01}$. It is possible that the charge builds up to a critical value and then escapes into the liquid.

(b) The fields in the vapor and the liquid will be different because the number density and mobility of the electrons in the two media are different and vary significantly with position.

(c) The alpha particles that ionize atoms in the vapor provide a source of electrons that is approximately independent of position in the region directly between the grids G0 and G1. However the alphas cannot reach all of the vapor region (see Fig. 3.1a) because they cannot pass though the 0.05 cm thick nickel disks that support the grids. Thus there is a lateral variation in the rate at which ionization occurs.

(d) The alpha particles that reach the liquid will ionize a large number of helium atoms within a distance from the surface of about 250 $\mu$m. Most of these will
recombine with the positive ions that are produced. The number of electrons that do not recombine depends strongly on the electric field in this region and based on the results of Williams and Stacey [77], the rate at which electrons are produced may be as large as the rate at which alphas enter the liquid.

In Fig. 5.14 we show plots of the measured signal at three different values of $\Delta V_{01}$ with the calculated leakage through the Frisch grid subtracted. Each plot includes the calculated signal based on the method just described, i.e., based on Eqns. 5.25 and 5.27 [78]. In each plot the calculated signal is shown for two values of $\epsilon_o$. The main discrepancy between the simulations and the data is seen in the time range between 90 and 100 % of the arrival time of the normal electron bubble; the simulation gives too large a result in this time range. It is interesting to consider the approximations made in the simulations. These include

(a) Equation 5.25 used for the energy distribution of electrons in the plasma.

(b) It is assumed that the bubble radius $R$ is proportional to $F^{1/4}$ and the mobility varies as $R^{-2}$.

(c) The potential barrier at the surface is assumed to be linearly proportional to the density as measured by Lurio et al. [75].

Of these assumptions c) seems to be the most questionable. It is not clear that for electrons of energy in the range of a few eV it is correct to view the helium as providing a potential which is smoothly varying on the length scale of the electron wavelength.

5.2.3.3 Testing the Leakage Model: Positive Ions

As a further test of the interpretation of the background signal and the calculation of the leakage, we have made a series of measurements for positive helium ions. A
Figure 5.14: The solid curve shows the measured #2 background after subtraction of the leakage current. The voltage difference $\Delta V_{01}$ between G0 and G1 is a) 20 V, b) 60 V, and c) 100 V. In each part of the figure the results for the calculated signal are shown for two different assumed values of the parameter $\epsilon_o$. 
positive helium ion $\text{He}^+$ moving in the vapor will lose a large fraction of its kinetic energy each time it collides with a neutral helium atom. Hence the typical energy of a helium ion incident on the surface of the liquid is of the order of $eE \Lambda$. From this we find that for most of the positive ions reaching the interface $\pi k_z a$ is large compared to unity. In addition, since the potential experienced by the ion in the liquid is negative relative to the vacuum, it follows that $\pi k'_z a$ is also large and greater than $\pi k_z a$. Thus from Eq. 5.27 the above the barrier reflection should be negligible and the only contribution to a background should arise from leakage through the Frisch grid. Hence a measurement of this background provides a test of the theory of the grid.

To make measurements with the positive ion we changed the sign of all the voltages applied in the cell. Typical results are shown in Fig. 5.15. The mobility is in reasonable agreement with previous measurements. An interesting feature of the results is the appearance of a bump in the data at a time which is about 7% larger than the arrival time of the main peak, and possibly another small signal arriving before the main peak. These features indicate the existence of ions $\text{He}^A$ and $\text{He}^B$ which have a lower and a higher mobility, respectively, than the normal $\text{He}^+$. We have been unable to find previous mention of these in the literature. An analysis of these objects follows in section 5.3.

Figure 5.16 shows plots of the positive ion signal on an expanded vertical scale and in the time range before the arrival of the main signal. The solid curve is the experimental data; the temperature is indicated in each plot. The measured signal has a ‘positive tail” extending out to times earlier than the main signal. However, the magnitude of the signal in this tail is much smaller than the background #2 signal for negative ions (see Fig. 5.9) and becomes lost in the noise at a time of about 75% of the arrival time of the main peak. To compare the positive tail with that expected from leakage through the Frisch grid we first integrated the current over
time to find the total charge $Q$ arriving per pulse of ions. This was found to have the values 0.069, 0.166 and 0.088 pC for the temperatures 1.044, 1.054, and 1.064 K respectively. We then made the approximation of treating the charge as a slab of zero thickness moving at the measured speed of the positive ion, and calculated the image charge, and the current due to the rate of change of the image charge with time. The results are shown by the dotted curves in Fig. 5.16 and are in good agreement with the measured “positive tail”. This indicates that unlike for the negative ions the only contribution to the background comes from the Frisch grid leakage.

In conclusion, we have described a method to trap a part of the wave function of an electron inside a bubble in liquid helium. We present evidence that even though the trapped wave function is in contact with the liquid this does not result in a measurement that determines that the electron is in or is not in the bubble. The experiments indicate that these electron bubbles have a lifetime of at least several tens of milliseconds.
Figure 5.16: Positive ion current arriving at the collector as a function of time for three temperatures. Solid curve shows the measured signal with the "positive tail" discussed in the text. The dotted curve shows the calculated signal arising from leakage of field lines through the Fisch grid.
5.3 Observations of New Positive Ions

As described in the previous section, measurements of the leading tail of the positive He$^+$ snowball were critical to verifying the theoretical model of the leakage signal and interpreting the #2 background. In the process of acquiring data for this purpose, we observed three peaks from positive charge carriers of discrete mobility, in addition to that from the He$^+$ snowball. In this section I will detail what we learned about how these new ions are generated, measurements of their mobility, and what these results might imply about their origins. These observations are still being analyzed, so this is simply an overview of what measurements were taken and where the analysis currently stands.

5.3.1 Summary of Previous Results

The idea of a dense charged helium core was first proposed by Atkins [15]. The electric field of the ion polarizes the nearby fluid, and the polarized helium atoms experience a strong attractive force as a result. Within a radius of about $\sim 7$ Å, this pressure is above the melting pressure, and so it converts the liquid into a hard core of approximately 40 helium atoms. The mobility of the He$^+$ snowball has been measured in a number of different experiments and found to be about twice that of the normal ion [12, 13, 79].

The Atkins’ argument applies reasonably well to other ions as well. A number of experiments have been designed to measure the mobilities of heavier ions. These experiments are performed by purposefully inserting impurities into the cell and then sputtering them at low temperature. The sputtered ions are then driven into a time of flight apparatus and collected so their mobilities can be ascertained. The first such experiments studying positive impurities were performed by Johnson and Glaberson [80, 81]. Their research team applied impurity laced solvents onto sharp tips. Once
dry, these tips were installed and used as discharge tips in the cell. When a high voltage was applied, impurity ions were sputtered off the tips and into the cell. The mobilities of the positive K\(^+\), Rb\(^+\), Cs\(^+\), Ca\(^+\), Sr\(^+\), and Ba\(^+\) ions were studied. A few decades later Foerste et al. [82] measured the mobilities of positive impurities Be\(^+\), Mg\(^+\), Ca\(^+\), Sr\(^+\), and Ba\(^+\) at temperatures around 1.5 K. The ions were sputtered as in the experiments of Johnson and Glaberson, but instead of applying a high voltage to a tip the light from a pulsed Nd:YAG laser was focused onto the frozen impurities. Kasimov et al. [83] were the first to measure the mobilities of negative impurity ions. Their experiment also used laser ablation to free frozen impurity ions rather than field emission. The mobilities of Cl\(^-\), F\(^-\), I\(^-\), Ba\(^-\), and Ga\(^-\) were measured. More recently, Krhapak [84] and Krhapak and Schmidt [85, 86] wrote a series of papers analyzing these existing mobility measurements of positive and negative impurities in the context of more detailed models of their structure.

5.3.2 Experimental Results

5.3.2.1 An Overview of the Discharge Conditions for Generating Positive Ions

A full cycle of assembling the cell, rebuilding the cryostat, preparing the apparatus, and cooling down the experiment takes at least two weeks. Because of this, once the experiment has been cooled, observations are typically made for at least a few consecutive days. We refer to a single cycle of this process as a ‘run’. The observations detailed here were made during three separate experimental runs.

As previously discussed, the ion generation and plasma mechanisms are notoriously complicated. The generation mechanisms show time dependence, meaning the relative strength of certain ions changes over the course of a run, and the voltages at which particular ions are produced change as well. The process of setting up a specific plasma configuration often shows hysteresis effects, meaning that how a
set of voltages is reached affects the final state of the plasma. What this means is that shifting voltages slowly towards one set of parameters sometimes results in a discharge with different characteristics from one reached by changing those voltages suddenly. To understand the total picture as well as possible we used a combination of techniques. In some cases voltages were adjusted only by small increments over a large range of values, to get a sense of how one parameter changes the discharge in a particular configuration. Because of the large number of adjustable parameters, it was also necessary to test major changes to one or more parameters at a time. Through this combination of approaches, we have been able to identify some general features of different discharge states. This is by no means exhaustive.

I will now summarize the three experimental runs and what was learned about the various voltage configurations in each. In almost all the cases detailed here, one signal peak had an amplitude much larger than the others. We measured the mobility of the ions contributing to this peak, and these values are in good agreement with those of the \( \text{He}^+ \) snowball mobility as measured by Schwartz [13]. Therefore, we refer to this ion as the \( \text{He}^+ \), and label the additional ions based on their mobilities relative to it.

- **Run 1**

  In the first of these experimental runs, we observed one additional positive ion in the reverse voltage configuration (REV I) and with the liquid level 1-2mm above G1. This ion has a mobility approximately 5\% larger than that of the normal \( \text{He}^+ \) snowball, and will be referred to as F1. F1 was well resolved and on the order of 10\% the strength of \( \text{He}^+ \). A signal recorded at \( T = 1.063 \) K showing the normal \( \text{He}^+ \) and F1 ions is in Fig. 5.17.

- **Run 2**

  In the second run, we discovered that there are two more positive ions, with
Figure 5.17: Trace recorded at $T = 1.063$ K in the reverse voltage configuration. The level of the liquid-vapor interface is approximately 1.0 mm above G1. The drift field 65 V/cm. The peak produced by the F1 ion is much smaller than the normal He$^+$ peak but still well resolved.

- Run 3

In the third and final experiment, we discovered two more unique discharge configurations. One can be considered a sort of parallel state to the reverse configuration, in which the plasma is on, the power supply voltages are the same, but there is a different vapor electrical behavior (REV II). In this state F1 is
Figure 5.18: Traces recorded at $T = 1.074$ K in the reverse and normal voltage configurations. The amplitudes of the He$^+$ peaks have been normalized to unity for clarity. The level of the liquid-vapor interface is approximately 1.0 mm above G1. The drift field 65 V/cm. S2 appears only as a tail in the normal configuration.

generated at large (i.e. 40pA) strengths, often larger than the He$^+$ strength. The second is a state in which the plasma is entirely off by keeping $V_S$ about equal to $V_{G0}$ and $V_{G0}$ only slightly more negative than $V_{G1}$ (OFF). Here, interestingly, F1 is generated on the order of 1pA with a normal He$^+$ amplitude on the order of 10 pA. Since there is no plasma in this state, and thus no sputtering from the walls of the cell, this indicates F1 does not originate from an impurity frozen on the cell.

Fig. 5.19 shows examples of the different voltage configurations as the voltage applied to the source is modified. The traces are grouped into three categories, the reverse configuration (0 V), the plasma off configuration (800 V), and the normal configuration (1000 V, 1500 V, 2000 V). All observations in Fig. 5.19 were made at a temperature of 1.084 K. The drift field was 65 V/cm.

In the first two experimental runs measuring the positive ions was not the primary focus, and they were only observed at the earliest $\sim 9$ hours after the discharge was
Figure 5.19: Traces recorded at $T = 1.084$ K in the REV II, OFF, and NORMAL voltage configurations. The level of the liquid-vapor interface is approximately 1.0 mm above G1. The drift field $65$ V/cm. F1 is strongest in REV II and comparable to the normal He$^+$ signal. It is substantially weaker in the OFF configuration when there is no plasma. The F1 signal is very weak in the NORMAL configuration, but the S1 ion becomes clear with increasing voltage applied to the source. Voltages in the legends indicate the voltage applied by the power supply to the source.
initially turned on. From there, data was taken over periods of days without any large changes in the signal characteristics. However, for the third run, positive ion data was recorded immediately upon completion of filling the cell. In this case the signal had an obvious time dependence. During the second run, the data were well resolved in the normal configuration at a source power supply voltage of 2000V. This time, during the first hour or so, data taken at about 1100V more closely resembled that from Run II, and at 2000V the signal was entirely absent. Over the course of the afternoon the lower threshold for seeing the positive ion increased substantially (a few hundred volts) and they began appearing at higher voltages (2000V for example). Taking repeated traces at the same setting also showed the pulse shapes changing. To summarize, in the normal configuration there is a voltage window over which a stable discharge is produced. There is initially a voltage at the lower end of this window where S1 is generated and well resolved. Over a time span on the order of hours this voltage increases at a decreasing rate, eventually settling to a seemingly stable value. Interestingly, this time scale is similar to that previously observed in experiments for the negative exotic ions. Whereas the exotic ions disappeared completely in these experiments, the additional positive ions have been observed for at least a week into experimental runs. The time dependence of the plasma and generation of negative exotic ions is currently a subject of research interest, and may be detailed in a future publication.

I would like to briefly point out that the F1 ion was possibly observed in a previous experiment by Wei et al. that is detailed in the thesis of Zhuolin Xie [9]. The left panel of Fig. 5.20 shows the positive ion data as the drift field is varied. This data was recorded by Wei et al. [8] and plotted in the thesis of Z. Xie [9]. The temperature was kept constant at 1.095 K. The right panel of Fig. 5.20 shows these same data, but where the arrival times of the normal He$^+$ ions have all been scaled to unity, and the integrals of the signals have also each been scaled to unity. In both panels two
'kinks' are visible in the signals, and are most pronounced for the observations at 35.5 V/cm. The kinked section of the traces cover times from approximately 90% to 95% of the normal He$^+$ arrival time. This is quite close to the relative arrival time we observe for F1 of $\sim$ 95-96%. Interestingly, these observations were made with sharp tungsten tips, indicating the type of source is not critical in generating the new ions.

5.3.2.2 Measurements of the Positive Ion Mobility

The positive ions were observed in the temperature range from 1.02 K to 1.24 K. As detailed in the previous section, there are numerous voltage configurations. In each used here G2 was held constant at +400V, and so the drift field was 65 V/cm. The gate pulse applied to G0 and G1 was of a constant +40V amplitude. The gate width was adjusted to optimize the signal peak for each unique parameter set. The cell was filled such that the liquid vapor interface was $\sim$ 0.5 mm above G1.

Drift velocities for F1 were obtained from the REV II configuration, in which the F1 and He$^+$ peaks are of similar size. G0 was held at +500V. In this configuration
the power supply for S is left off. A plasma forms between S and G0, resulting in a measured voltage of approximately +300V on S. This measured voltage on S is much higher than that in the more easily accessible primary reverse configuration (~200V). There was no foolproof method of producing the REV II configuration. Typically it was done by arranging the normal voltage configuration, and then suddenly switching off the power supply for S. This, however, often took multiple attempts and would fail except at the low end of the temperature range. When it did fail, the REV I configuration was produced.

For S1, data taken in the NORMAL configuration was used. Here G1 was maintained at +420V. The voltage on S was held between +550-570V. In Fig. 5.21 we see that the relative mobilities of the F1 and S1 ions only change very slightly over the temperature range. Linear fits of the F1 and S1 relative mobilities give slopes for each of -0.043/K, corresponding to a 1% change in the mobilities over the range measured. The average mobility for F1 and S1 relative to the He$^+$ snowball mobility, are found to be 105% and 94%, respectively.

S2, however, is small and convoluted with S1. For each trace we fit a sum of three Gaussian functions to obtain the S2 arrival time, but the results showed a far larger variance than either the F1 or S1 ions. This is likely due to the difficult nature of accurately fitting a peak that appears as a small kink. Due to a lack of confidence in this procedure at the current time, observations of S2 are not included in this or other plots.

Glaberson and Johnson [81] observed a temperature dependent shift as well, but differing in important ways. They found that the impurity ions they studied formed two groups, Rb$^+$, Cs$^+$, and K$^+$, with mobilities smaller than the normal He$^+$ ion, and Ca$^+$, Sr$^+$, and Ba$^+$, with mobilities exceeding it. Regardless of the impurity species, they found that the mobilities always approached the normal He$^+$ ion mobility as the temperature was increased (1.1 K to 1.5 K). These observed shifts were also quite
Figure 5.21: Mobilities of the F1 and S1 ions relative to that of the normal He$^+$ ion. Temperature was adjusted from 1.022 K to 1.238 K. Mobilities were calculated for the F1 ion from data taken in the REV II configuration, and for the S1 ion the NORMAL configuration.

large compared to that which we’ve observed here, anywhere from 1-20% over 50 mK depending on the species.

Next we searched for any critical values for the ion velocities. Discrete shifts in the peak amplitudes would be of particular interest, as they would speak to structural stability of the clusters. The drift field was varied in strength from 49 V/cm to 130 V/cm. This is a small range, but the design of our experimental cell, most critically the electrical feedthroughs and the close proximity of electrodes which are not submerged in the liquid, create strict limitations. The field dependent mobilities of F1 and S1 are shown in Fig. 5.22. Although there does appear to be a small continuous downward slope in the mobilities, we have not observed any sudden shifts indicating changes to the cluster structure.

5.3.2.3 Measurements of the Positive Ion Amplitudes

Using the same datasets detailed in the previous section for obtaining the mobilities, the amplitudes of F1 and S1 relative to He$^+$ were calculated. Here the amplitude
Figure 5.22: Mobilities of the F1 and S1 ions relative to that of the normal He$^+$ ion. The drift field was varied from 48.8 V/cm to 130.1 V/cm. Mobilities were calculated for the F1 ion from data taken in the REV II configuration, and for the S1 ion the NORMAL configuration.

is defined as the peak of the signal. These results are illustrated in Fig. 5.23. The discrete shifts in the S1 amplitude correlate with adjustments to the gate pulse width.

5.3.2.4 Effect of Liquid Interface Level

Previous measurements of the ion mobilities have found a significant dependence on the position of the helium liquid-vapor interface [5, 21, 23]. The liquid height is one of the more difficult parameters to vary in this experiment. In our cell, filling or removing helium is only possible by pressurizing or pumping the helium fill lines, and so both operations necessarily induce large heat flows. As a result, temperature shifts occur during these operations, and this precludes maintaining the discharge condition. These are also time consuming processes, and so they are often avoided during runs in favor of other observations that can be made more readily. This all to say that while observations were made during Run I of the effects of the liquid height on the F1 ion, they were not in great detail nor were follow up observations.
Figure 5.23: Amplitudes of the S1 and F1 ions relative to that of the normal He\textsuperscript{+} ion. The temperature was varied from 1.022 K to 1.238 K. Mobilities were calculated for the F1 ion from data taken in the REV II configuration, and for the S1 ion the NORMAL configuration.

made during the Runs II or III. It also means that any time dependent behavior of the discharge is difficult to separate out from effects of the interface height.

In Fig. 5.24 the peaks of the F1 and normal He\textsuperscript{+} ions at three different liquid levels, 1.0 mm, 2.0 mm, and 4.0 mm above G1 are shown. In each trace the peak of the normal He\textsuperscript{+} ion is normalized to unity. It is clear that the F1 ion which was well resolved at 1.0 mm loses strength relative to the normal He\textsuperscript{+} ion as the height is raised.

5.3.2.5 Interpretation

The mobility of each ion should be proportional to the inverse cross section of the ion,

\[ \mu_i(T) = A_i(T) \frac{1}{R_i^2} \]  \hspace{1cm} (5.28)

where \( A_i \) is a coefficient accounting for the temperature dependent roton scatter-
Figure 5.24: Traces recorded at $T = 1.063$ K in the reverse voltage configuration. The amplitudes of the He$^+$ peaks have been normalized to unity. The height of the liquid-vapor interface was varied between 1.0 mm and 4.0 mm above G1. The drift field 65 V/cm. The F1 peak gets noticeably smaller with increasing height of the interface.

If we use the same procedure described in section 5.1 for estimating the size of the negative bubbles, and assume the radius of the normal He$^+$ snowball is 7 Å, the radii of these additional positive ions can be estimated. Additionally, if $N_o$ is the equilibrium number of helium atoms in an ionic cluster, and we assume each helium atom of the cluster occupies an equal volume, then for a cluster of mobility $\mu'$ we can estimate the number of helium atoms in it by,

$$\frac{\mu'}{\mu_o} = \left( \frac{R_o}{R'} \right)^2 = \left( \frac{N_o}{N'} \right)^{2/3}$$

(5.29)

The mobilities of the additional positive ions F1 and S1 are approximately 105% and 94% of the He$^+$ mobility, respectively. This would correspond to radii of 6.8 Å and 7.2 Å. The changes to the number of atoms in the cluster are estimated to be $\Delta N_{F1} = -3$ and $\Delta N_{S1} = +4$. An interesting possibility, but one we have not explored in depth, is that the He$^+$ ion forms more than one stable configuration in the liquid. Changing the geometric configuration of the neutral helium atoms in the
cluster would modify the helium density near to the ion, and thus the number of atoms within the radius predicted by Atkins.
Bibliography


[33] Private communication from Paul Leiderer.


[50] EKSPLA, Savanoriu Avenue 237, LT-02300 Vilnius, Lithuania.


Note the following interesting property of a Frisch grid. For a suitable choice of the geometry, the grid can be made to have a small leakage of field lines coming from approaching ions. But for this same geometry most of the field lines from the electrodes in the cell that provide the \textit{static} drift field will pass through the grid. As a result most of the ions pass through the grid rather than being captured by the grid wires. For a discussion see O. Buneman, T.E. Cranshaw, and J.A. Harvey, Can. J. Res. A\textbf{27}, 191 (1949). For our grid we have calculated that \(~88\%\) of the static field lines ions pass through the grid. Thus assuming
that the ions follow the field lines only $\sim 12\%$ of the ions are lost at the Frisch grid.


[73] See, for example, M.J. Druyvesteyn and F.M. Penning, Rev. Mod. Phys. 12, 88 (1940).


[78] As previously mentioned, the normal bubbles are treated as an infinitesimal slab for the purposes of the leakage calculation. To test the validity of this simplification, we fit the NB peaks with Gaussian functions, and then simulated the leakage current produced by these Gaussian shaped slabs as they travel. There was a negligible difference between the two methods, and so we continued to model the slab as thin sheet of charge.


