# Atomic Hydrogen Cleaning of Polarized Electron Source Gallium-Arsenide Photocathodes

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by

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## ABSTRACT

Within the past few decades, particle accelerators have become required tools in nuclear physics research. Polarized electron sources are one of the many important aspects of these tools at certain accelerator facilities, and have been put to use in high-energy physics for the studies of nuclei and quarks. Current high-energy polarized electron sources utilize photo-stimulated emission from a gallium-arsenide cathode. One of the many difficulties in the creation of high-current, highly-polarized electron beams comes from the contamination of the cathode itself, which eventually decays the output electron beam current to unusable levels. Recent research focuses on an atomic hydrogen cleaning process that has the ability to remove contaminants from the cathode's surface more effectively and efficiently than previous methods. This thesis will seek to quantify the method of this atomic hydrogen cleaning process in an effort to maximize its effectiveness, thereby increasing the overall cathode lifetimes.

### I. INTRODUCTION

Highly polarized electron beams are becoming crucial research tools in modern high-energy nuclear physics. One of the few high-energy polarized electron beams available to the physics research community exists at the Thomas Jefferson National Accelerator Facility. Polarized electron creation is accomplished in an ultra-high vacuum environmentusing photoelectric emission from a dime-sized gallium-arsenide wa fer. This photoelectric emission is stimulated by circularly-polarized infrared laser light incident on the GaAs cathode. The ultra-high vacuum chamber in which this photoemission takes place is kept at pressures on the order of 10<sup>-10</sup> torr to provide a clear path for the electron beam. Once free from the surface, polarized electrons accelerate away from the negative electron affinity cathode across 100kV beyond which they are focused for their subsequent injection into the main accelerator. The Polarized Electron Source Lab at TJNAF performs continuing research to improve overall electron emission and polarization.

One of the many difficulties in polarized electron beam creation occurs at the gallium-arsenide crystal itself. The quantum efficiency of GaAs is highest at photostimulative wavelengths around 530nm. However, in order to extract highly polarized electrons from the surface, circularly-polarized light in the infrared ranges must be utilized.<sup>7</sup> Unfortunately, GaAs has a very low quantum efficiency for photoemission at the infrared wavelengths. Although it is possible to obtain high polarized beam at the high currents necessary to perform the high-energy nuclear physics for which Jefferson Lab is so well known.



The chamber shown in Figure 1 illustrates the layout of the polarized source electron gun. The left side of the figure shows the high voltage components that are held at a negative 100kV potential as well as the surrounding insulating ceramic. The GaAs cathode is mounted at the end of the stalk that extends lengthwise into the chamber left to right. A chemical NEG pump array surrounds the cathode to keep the source as contaminant-free as possible. The right portion of the figure shows the cesiator that retracts during beam operation to allow the electron beam free passage into the injector to the right side of the diagram.

First-generation electron guns stood upright with the infrared laser incident from underneath the gun through an ultra-high vacuum window. The electron beam itself was

produced perpendicular to the ground but was then steered 90° using a dipole magnet to align it parallel to the ground for injection into the accelerator. Current polarized sources lay horizontally to eliminate the need for a 90° dipole magnet, but the incident infrared laser beam is now inserted through an ultra-high vacuum window on the side of the chamber and reflected onto the catho de within the vacuum off a highly-polished stainlesssteel mirror.

Because the GaAs cathode is required to operate at such low quantum efficiencies, other effects that in optimal situations would have only small influences, in fact have quite a large impact on the quantum efficiency. The most influential of these effects is surface contamination of the cathode.

To reduce the photoelectric work function of the crystal, its surface is coated with a monolayer of cesium oxidized by oxygen or nitrogen trifluoride. The work function at the surface is particularly sensitive to changes at the boundary layer so keeping the crystal contaminant-free is extraordinarily important. In the past, efforts to keep the GaAs clean included a chemical etching process as a vacuum preparation technique.<sup>2</sup> This process yielded somewhat unpredictable results on the molecular level in the cleanliness of any given wafer and has since been abandoned. Today, photocathodes are 'etched' under vacuum using an atomic hydrogen technique, but are then exposed to atmosphere for transport to the polarized source. Once the wafer is under vacuum in the polarized source, the entire vacuum chamber is baked out at 300°C for a 24 hour period in an effort to liberate most of the residual gases trapped in the stainless steel chamber walls. This bakeout process yields orders of magnitude lower base pressures for the polarized source chamber, consequently permitting a clear path for the electron beam as it is created and injected into the accelerator. After the chamber bakeout, the GaAs wafer itself is heated to 600-700°C in order to liberate any contaminants on the surface including the residual gases absorbed at atmosphere.<sup>2</sup> Finally, to create a cathode, the GaAs wafer is coated by a monolayer of cesium as the last step in the long process of preparing to deliver polarized beam to the injector. Unfortunately even with excruciating care taken to keep the GaAs as sterile as possible, there are still possibilities for contamination.

One unavoidable type of contamination is a direct byproduct of electron beam production. Once the source is in operation, the electron beam itself ionizes residual gas molecules which then accelerate toward, and build up on, the gallium-arsenide cathode, which is held at a potential of negative 100kV. This contamination layer, which builds up even at pressures as low as 10<sup>-10</sup> torr, decreases the quantum efficiency of the surface substantially for a given incident laser intensity, thereby reducing the output beam current.<sup>7</sup> Only by decreasing the ultra high vacuum pressure orders of magnitude further could this problem be completely eliminated. The expense in equipment and labor to lower the polarized source vacuum pressure to the order of 10<sup>-13</sup> torr would be enormous, requiring another solution to the problem. The only currently feasible solution is to develop an effective way of cleaning the cathode while it is still in the gun.

Once a gallium-arsenide crystal is covered with multiple monolayers of ionized gas, its quantum efficiency drops below a usable level. At this point, the wafer must either

be exchanged or cleaned. It is possible to reheat the wafer in order to liberate the unwanted surface contamination and then re-cesiate, but this method can only temporarily extend the lifetime of the cathode. A method of reliably cleaning the cathode without venting the chamber to atmosphere (which requires days of downtime to recover an ultrahigh vacuum environment) is needed for the continuous beam delivery demanded by Jefferson Lab's users.

Further difficulties include cleaning of new 'strained' GaAs cathodes that have recently become the standard for polarized electron emission. These cathodes are too thin (~100 nm) to withstand a wet chemical etch process.<sup>3</sup> Research continues to improve methods of cathode cleaning that will avoid excessive downtime and will yield more uniformly scrubbed cathodes. The atomic hydrogen cleaning process lies on the cutting edge of that research.

### II. THEORY

Current research involves a hydrogen source that releases atomic hydrogen into the chamber in an attempt to clean the wafer while still under high vacuum conditions. In the atomic state, hydrogen and its isotopes have very high affinity to bond and will readily do so with contaminants in the chamber. By bombarding the GaAs wafer with atomic hydrogen, electronically-bound contaminants on the surface are more likely to bond with the hydrogen and thus are liberated from the surface. These contaminants, as well as the

hydrogen to which they are now bound, are then pumped away. Of course, because of atomic hydrogen's affinity to be bound, not all of the atomic hydrogen can be used for cleaning as it will recombine into its lower-energy molecular state. Thus, to maximize the cleaning ability of the atomic hydrogen source, the amount of atomic hydrogen that leaves the dissociator per unit time must also be maximized to allow for the most thorough atomic bombardment of the surface.

The atomic hydrogen source itself consists of a molecular hydrogen dissociator made up of a glassware vacuum component (in which the dissociation occurs) surrounded by a wire coil that rests inside of a grounded brass cylinder.<sup>1,6</sup> The coil is a part of an LC tuned circuit that resonates at approximately95 Mhz. This resonant RF energy breaks the molecular bond and dissociates the hydrogen into atoms. Upon dissociation, the atomic hydrogen flows through a small 1.0mm aperture into the chamber where it is then used for cleaning. This atomic hydrogen system uses deuterium because it is more easily pumped away. Although preliminary attempts at this hydrogen cleaning process seem to yield successful results, quantification of the cleaning process and the dissociator's operational parameters is quite difficult.

The hydrogen dissociator has two operational parameters which can be used to maximize the atomic output. First, the quantity of molecular deuterium introduced to the dissociator can be varied to determine what input pressure of deuterium gas will yield a maximum atomic output. At low input pressures, less than 10 mTorr in the dissociator, not enough molecular deuterium is available to maintain a steady-state dissociation. At

input pressures in excess of 50 mTorr in the dissociator, recombination of the atomic deuterium is prevalent due to such large numbers of free atoms. The second operational parameter involves varying the input RF to determine what absorbed power optimizes dissociation performance. The resonant frequency is determined theoretically and does not need experimental verification, but it is potentially useful to determine the optimum RF power absorption for a given dissociator input pressure as absorbed power varies with the amount of deuterium in the dissociator.

The initial successes of this atomic hydrogen cleaning process have prompted the replacement of the wet chemical etch process, previously used to initially prepare GaAs cathodes, with hydrogen cleaning. Currently, wafers are atomic hydrogen cleaned in a separate vacuum chamber that is vented to nitrogen after the cleaning process. This exposure to atmosphere seems to be fairly benign as the duration is short and the wafer only sees pure nitrogen gas during its transport to the polarized source. Nitrogen is easily cleaned off the surface once the wafer is under vacuum through the heat treating process that prepares the GaAs wafer for cesiation. Although this hydrogen cleaning process has seen success, the operational parameters of the atomic hydrogen source have never been quantified.

The purpose of the thesis is to quantify those operational parameters in an attempt to maximize atomic output and therefore maximize cleaning ability of the hydrogen dissociator. This requires the construction of an experimental vacuum chamber with the ability to determine atomic deuterium output.

### III. THE EXPERIMENTAL SETUP

The experimental setup consisted of a differentially pumped vacuum chamber that was placed on the existing dissociation chamber. The chamber was constructed of three five-way vacuum crosses to allow for the mounting of pumps and other necessary equipment. There were three subsections, see Figure 3, in which the differential pumping was accomplished using gaskets between the subsections with a measured aperture of

exposed directly to the was dissociator output and had a 70 L/s turbo pump and a 20 L/s ion pump. The turbo pump ran continuously while the dissociator was in operation due to its relative efficiency in pumping low-mass residuals such as hydrogen. Since the advertised pump rates on ion pumps are determined using species that are more easily ionized than hydrogen, such as nitrogen, the ion

10mm. The lowest of these sections



pumps on the chamber had difficulty pumping such large amounts of mass two and mass

four because hydrogen is so difficult to ionize. The middle section initially had a 30 L/s ion pump and was used simply to enhance the differential pumping between the top and bottom sections. The top section of the chamber had a 20 L/s ion pump. This section housed the residual gas analyzer (RGA) mounted line-of-sight through the apertures to the dissociator. The RGA measured the partial pressure of deuterium in the upper section of the chamber where the pressure was low est.

This differential pumping system was necessary for two reasons. First, RGAs will not operate at pressures in excess of 10<sup>-6</sup> torr which were common in the section directly exposed to the dissociator. Second, it was necessary to pump away as much of the background as possible in order for the RGA to see line-of-sight to the dissociator. The upper section also contained the shutter that could selectively block or open the line-ofsight path from the dissociator to the RGA in order to accurately measure the background residual gas.

This shutter was constructed on a rotating feedthrough mounted perpendicular to the line-of-sight between the dissociator and the RGA. The shutter itself consisted of an aluminum three-sided cube structure that was mounted to the feedthrough on the middle side. This structure was



Figure 4. Line-of-sight shutter

mounted perfectly centered in the chamber so that when the shutter was open the dissociator would be visible to the RGA between the two sides of the shutter structure. By rotating the feedthrough 90°, this structure would obstruct the RGA's view of the centerline aperture below the shutter. The construction of this shutter created a method of subtracting out the background partial pressures of mass four that became visible to the RGA once the dissociator was in operation. An accurate measure of the mass four background with the deuterium flowing into the dissociator with both RF on and off was necessary in order to observe only the line-of-sight output from the dissociator to the RGA.

### IV. THE EXPERIMENTAL PROCEDURE

Operational parameters in the setup are varied in order to maximize the output of atomic deuterium, which is quantified using the dissociation fraction, D, defined by Equation 1. This equation requires a measurement of the quantity of molecular deuterium (mass four) in the chamber coming directly from the dissociator, given by N. Molecular

$$D = \frac{N^{RFoff} - N^{RFon}}{N^{RFoff}}$$

#### **Equation 1.**

deuterium, not atomic deuterium, is measured exclusively because of the experimental

inaccuracies in determining the partial pressures of species with masses close to zero. Data acquisition requires a molecular deuterium flow into the dissociator while mass four measurements are taken with both the RF on and off, N<sup>RFon</sup> and N<sup>RFoff</sup> respectively. The dissociation fraction is then determined by taking the difference between the mass four signal RF on and off, which corresponds to the quantity of dissociated hydrogen, over the total molecular hydrogen output of the dissociator. The difficulty in this measurement arises in the inability to measure only dissociator output without also seeing extraneous mass four background that has not traveled line-of-sight from the dissociator.

To solve this problem the line-of-sight from the dissociator output must be selectively blocked in order to accurately measure the mass four background. This requires four different measurements of the mass four signal: RF on and off with the dissociator output open and RF on and off with the dissociator output blocked. Now equation 1 can be rewritten as

$$\frac{(A-B)-(C-D)}{(A-B)}=D$$

### **Equation 2.**

where **A** and **B** are the mass four signals RF off with the dissociator open and blocked, respectively, and **C** and **D** are the mass four signals RF on with the dissociator open and blocked respectively.



Figure 2. Expected mass four signals for a 50% dissociation fraction

Figure 2 follows the same convention as Equation 2 and shows a theoretical set of mass four signals that would yield a 50% dissociation fraction. This dissociation fraction can then be maximized using the operational parameters of the hydrogen dissociator.

The output max imization of atomic deuterium from the dissociator will yield the maximum effectiveness in the atomic hydrogen cleaning process of GaAs photocathodes. This optimization is becoming more and more necessary as photocathodes are now being cleaned using this atomic hydrogen bombardment technique. Cleaner, more uniform cathodes, will yield longer operational lifetimes and improved quantum efficiency which

will eliminate the necessity to swap out cathodes as often as is currently required.

### V. RESULTS

This initial experimental setup yielded no usable measurement of the dissociation fraction for a number of reasons. The first and foremost was the poor differential pumping ability of the chamber which led to the inability of the RGA to see a line-of-sight mass four signal from the dissociator. The pressures between the individual sections were only lower by a factor of two as opposed to the desired differential pumping of close to an order of magnitude between each section. Attempts to solve this problem were two-fold. First, the aperture size between each section was reduced to 5mm thereby limiting the conductance section-to-section by a factor of four. Second, a 200 L/s chemical NEG pump was added to each of the top two sections of the chamber in order to further reduce the background. The NEG pumps used had been exposed to atmosphere for quite sometime and almost certainly did not pump at their advertised rate. Nonetheless, after they were activated they did help improve the overall differential pumping within the chamber. Chart 1 shows the differential pumping ability of the chamber before and after these improvements for a given dissociator pressure.

| Upper      | 0.43          | 0.19         | 0.03          | 0.02           |
|------------|---------------|--------------|---------------|----------------|
| Middle     | 0.93          | 0.36         | 0.12          | 0.04           |
| Lower      | 1.00          | 1.00         | 1.00          | 1.00           |
| Subsection | 10mm aperture | 5mm aperture | 5 mm aperture | 2.5mm aperture |
|            | (NEGs off)    | (NEGs off)   | (NEGs on)     | (NEGs on)      |

Differential Pumping Ratios at 25 mTorr

#### Chart 1.

A 2.5mm diameter aperture was also tested but without success. Although the differential pumping markedly improved, as the conduction between sections was once again reduced by a factor of four, the aperture appeared to block the line of sight from the RGA to the dissociator.

The dissociator itself has a 1.0mm output aperture which creates two experimental difficulties. First, it is difficult to determine whether the hand-blown dissociator has its aperture perfectly centered along the vertical axis of the chamber. Without this alignment it would be impossible for the RGA to see the dissociator output directly through the two inter-chamber apertures, particularly with apertures as small as 2.5mm. Second, over the distance traveled by the deuterium to the RGA, openings of 2.5mm can block the edges of the line-of-sight mass four signal and that would have otherwise been visible to the RGA.

One other major improvement in an attempt to attain usable data was the relocation of the line-of-sight shutter in the chamber. With the shutter located in the uppermost section of the chamber, it was ineffective in blocking the line-of-sight mass

four signal, although it would block the mass two signal. Blocking of a mass two signal is mostly attributed to the mass four background being so high that the pumps in the upper chamber did not remove the mass four before it made its way around the closed shutter to the RGA. The shutter did block the mass two signal though because after the line-ofsight mass two bounced off the closed shutter it would recombine into mass four before being detected by the RGA. Although this mass two effect was visible with the RGA, no valid data was taken with the mass two signal because of known errors in mass two RGA measurements due to the species' low mass.



Figure 5. Mass four signals with the shutter mounted in the upper chamber

Figure 5 depicts some of the curious mass four signals with the shutter in the upper chamber. **A** should yield the largest amount of mass four as it is RF off with the shutter open. **B** would be expected to exhibit a smaller mass four signal than **A** since some of the mass four should be blocked by the shutter. Seeing the same mass four signal for both **A**  and **B** indicates the shutters inability to block any of the mass four coming from the dissociator. **C** shows an order of magnitude larger mass four signal which can be attributed to the dissociator causing some type of RF coupling with the RGA and therefore creating an ambiguous mass four signal. The possibility also exists for the recombination of atomic deuterium within the RGA head before it is analyzed which could artificially boost the mass four signal (although not more than the RF off mass four signal). Unfortunately, neither of these causes are easily rectified. The mass four signal RF on with the shutter closed, **D**, seems to be reasonable but this combination of signals does not yield a dissociation fraction.

Another reason for this difficulty in detecting a reliable line-of-sight mass four signal is believed to be partially due to the relatively high pressures in the lower chamber that partially dispersed the mass four signal enough to preclude its accurate measurement. Without a good measurement of the mass four coming directly out of the dissociator, a reasonable dissociation fraction measurement is impossible. In an attempt to rectify this, it was decided that the shutter would be relocated to the bottom chamber directly above the dissociator. This shutter location led to a measurable difference in the mass four signal with the shutter open and closed because of the shutter's ability to block the mass four signal before it became background that was visible to the RGA. This shutter location kept most of the mass four blocked by the shutter from making its way to the RGA by keeping the blocked mass four in the lower chamber. Unfortunately, this setup still led to no usable data, although this data was closer to



Figure 6. Mass four signals with the shutter mounted in the lower subsection

expected data than with the shutter in the upper chamber.

Figure 6 shows a sampling of data taken with the lower shutter position. The shutter now has an effect on the RF off mass four signal but the difference is smaller than the RF on mass four signal. This leads to an impossible dissociation fraction greater than 100%.

With some of the more extreme variations in the operational parameters of the dissociator (where the dissociation fraction is expected to be low), this setup did yield fractions less than 100%. Unfortunately, most of this data was virtually impossible to replicate, forcing the conclusion that the experimental errors in the system were too large to yield useful data.

## VI. CONCLUSION

It is clear that a pure line-of-sight mass four signal from the dissociator was not being detected by the RGA. It is thought that the background in the chamber still remains too high to see only a line-of-sight signal. Reduction of the background to a level where much less stray mass four is detected should result in a more accurate RGA measurement. The most effective way to ensure a small background mass four signal is to substantially increase the overall pumping on the chamber. Large pumps in excess of 500L/s on each chamber would most likelytake care of the mass four background problem and allow the RGA a clear view of dissociator output. This 'brute force' approach may be the only way to ensure a true line-of-sight mass four signal.

Shutter position is the other critical aspect of experimental operation. The most effective shutter location for a good line-of-sight mass four signal is within as close a proximity as possible of the dissociator without compromising its output ability. This location allows for the best blocking of the line-of-sight signal while still leaving the background signal intact for accurate measurement.

In conclusion, the experimental process of attempting to determine dissociation fraction as a function of dissociator parameters proved to be a daunting task. But despite these difficulties, atomic hydrogen cleaning of gallium-arsenide photocathodes continues to be an extremely successful process which can only be improved upon by ongoing research into atomic hydrogen output maximization. Further research into this cleaning process will certainly yield the long-term benefits of cleaner photocathodes with longer operational lifetimes.

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