

IN-SITU CLEANING OF METAL CATHODES USING A HYDROGEN ION BEAM

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Abstract

Metal photocathodes are commonly used in high-field RF guns because they are robust, straightforward to implement and tolerate relatively poor vacuum compared to semi-conductor cathodes. However these cathodes have low quantum efficiency (QE) even at UV wavelengths, and still require some form of cleaning after installation in the gun. A commonly used process for improving the QE is laser cleaning. In this technique the UV drive laser is focused to a small diameter close to the metal's damage threshold and then moved across the surface to remove contaminants. This method does improve the QE, but can produce non-uniform emission and potentially damage the cathode. Ideally an alternative process which produces an atomically clean, but unaltered surface is needed. In this paper we explore using a hydrogen ion (H-ion) beam to clean a copper cathode. We describe QE measurements over the wavelength range of interest as a function of integrated exposure to an H-ion beam. We also describe the data analysis to obtain the work function and derive a formula of the QE for metal cathodes. Our measured work function for the cleaned sample is in good agreement with published values, and the theoretical QE as a function of photon wavelength is in excellent agreement with the cleaned copper experimental results. Finally, we propose an in-situ installation of an H-ion gun compatible with existing s-band RF guns.

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INTRODUCTION AND DESCRIPTION OF THE EXPERIMENT

Improving and maintaining the quantum efficiency (QE) of a metal photocathode in an s-band RF gun requires a process for cleaning the surface. In this type of gun, the cathode is typically installed and the system is vacuum baked to ~200degrees C. If the QE is too low, the cathode is usually cleaned with the UV-drive laser. While laser cleaning does increase the cathode QE, it requires fluences close to the damage threshold and rastering the small diameter beam, both of which can produce non-uniform electron emission and potentially damage the cathode.

This paper investigates the efficacy of using a low-energy hydrogen ion beam to produce high-QE metal cathodes. Measurements of the QE vs. wavelength, surface contaminants using x-ray photoelectron spectroscopy and surface roughness were performed on a copper sample, and the results showed a significant increase in QE after cleaning with a 1keV hydrogen ion beam. The H-ion beam cleaned an area approximately 1cm in diameter and had no effect on the surface roughness while significantly increasing the QE. The work function for copper is extracted from this data. In addition, a formula of the QE for metals as a function of optical wavelength is derived assuming the 3-step process for photo-emission and using the Fermi-Dirac distribution for the electron density of states (EDOS). The theory is in excellent agreement with measurements of QE for a clean copper surface. These results and a method for installing an H-ion cleaner on existing s-band guns are described.

Sample Preparation and Surface Roughness

A sample 25mm in diameter and 2.5mm thick was polished using 0.25micron diamond paste. However, during its storage under hexane, the hexane had evaporated resulting in an oxidized carbon surface and a very low initial QE (Figure 1). The surface roughness was studied to determine if there was any change in the surface morphology due to the exposure to the H-ion beam. Atomic force microscope (AFM) measurements on a similarly treated sample gave the following results [1]. The Ra and Rq before treatment were 10.55nm and 13.09nm respectively, and the peak-to-peak roughness was 187.91nm. After exposure to H-ions the respective values were 12.48nm, 15.65nm and 118.76nm for one 50x50micron area and 10.19, 12.52 and 82.50nm for another 50x50micron area. Two areas were measured in order to estimate the systematic uncertainty of not being able to repositioning the probe in exactly the same region on the sample when removed and then reinstalled in the AFM.

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QE and Surface Contamination Measurements

After preparation the sample was placed in a load lock chamber separated by a valve from an X-ray Photoelectron Spectroscopy (XPS) measurement chamber. The XPS data provided the percentage coverage of carbon on the surface. The QE measurements and the H-ion cleaning were performed in the load lock chamber. The QE was determined with the sample biased at -18V relative to the surrounding chamber. This was done to ensure that no secondary electrons generated by the chamber walls could return to the sample. Since there was a large gap (of order centimeters) to the nearest ground, the electric field on the sample was essentially zero. The sample was illuminated by light from a xenon arc lamp whose output was selected by a monochromator with a slit set to pass a wavelength range of 8nm. The transmission of the vacuum window and other optics were measured and used to determine the absolute QE. The H-ion beam was produced by a commercial saddle field ion source [2], whose beam energy was approximately 1KeV. Ion currents of 0.4, 0.7 and 2microamperes were used. Labview [3] software automatically scanned the wavelength in 2nm steps while collecting the photo- and background currents. The experiment determined the QE and the surface contamination as the sample was progressively exposed to the H-ion beam.

The data for the unbaked copper sample are shown in Figure 1. In other experiments it is standard practice to bake the sample to 230degrees C to clean the surface. In this case however, we were interested in the effects of H-ion cleaning only, therefore the initial bake was not performed. The H-ion charge is measured by integrating the sample current, which may include a contribution from charged contaminants leaving the surface.

The results are quite dramatic, especially at the drive laser wavelengths of interest at 255nm and 263nm. Although the sample was exposed to a total integrated H-ion charge of 10.23millicoulomb (mC), most of the benefit was achieved by 3.03mC. It should be noted that 250μJ at 255nm requires a QE of 2×10^{-5} to produce 1nC of electrons.

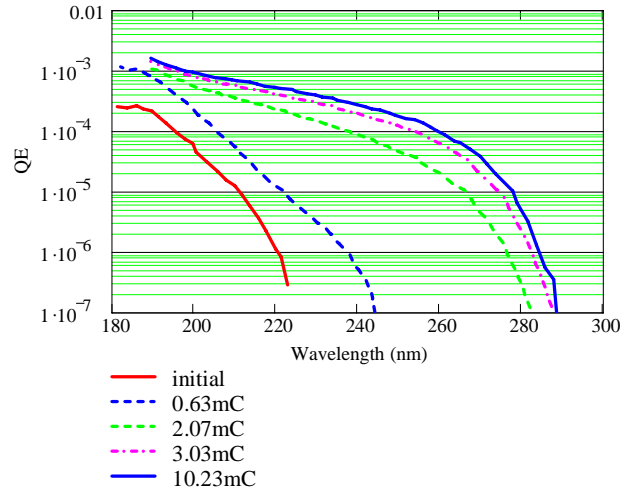


Figure 1: The QE for a copper sample vs. wavelength with increasing integrated exposure to the H-ion beam.

DATA ANALYSIS

The QE data were analyzed to obtain the work function using the method of Fowler [4]. This technique includes the effect of temperature at photoemission threshold by assuming a Fermi distribution for the electrons and comparing the data with the following function,

$$\ln\left(\frac{QE}{T^2}\right) = B + \ln\left(f\left(\frac{\hbar\omega - \phi}{k_B T}\right)\right) \quad [1]$$

where T is the electron temperature (assumed to be 300degK), k_B is Boltzman's constant, $\hbar\omega$ is the photon energy, and ϕ is the work function. B is a constant related to the electron density of states, the optical reflectivity and electron transport to the surface. The function $f(x)$ results from integrals of the Fermi-Dirac function and is approximated by

$$f(x) = e^x - \frac{e^{2x}}{4} + \frac{e^{3x}}{9} - \dots \quad \text{for } x \leq 0 \quad [2]$$

$$f(x) = \frac{\pi^2}{6} + \frac{x^2}{2} - \left(e^{-x} - \frac{e^{-2x}}{4} + \frac{e^{-3x}}{9} - \dots\right) \quad \text{for } x \geq 0 \quad [3]$$

Fowler showed that ϕ and B are easily obtained by plotting the experimental $\ln(QE/T^2)$ vs. the photon energy normalized to $k_B T$, and fitting with $B + \ln(f((\hbar\omega - \phi)/k_B T))$. Fowler plots are given for our six QE data sets in Figures 2, 3 and 4 and the percentage of carbon coverage is listed above each plot. The comparison with Fowler's theory is

excellent with the exception of few cases. Figure 2 shows the data and theory for the initial, contaminated sample with 31% carbon coverage and after a short exposure to the H-ion beam (0.630mC). In these cases the fit has been biased to better match the threshold region. We assume the discrepancy at higher photon energies results from additional electron states not accounted for by the Fermi-Dirac distribution.

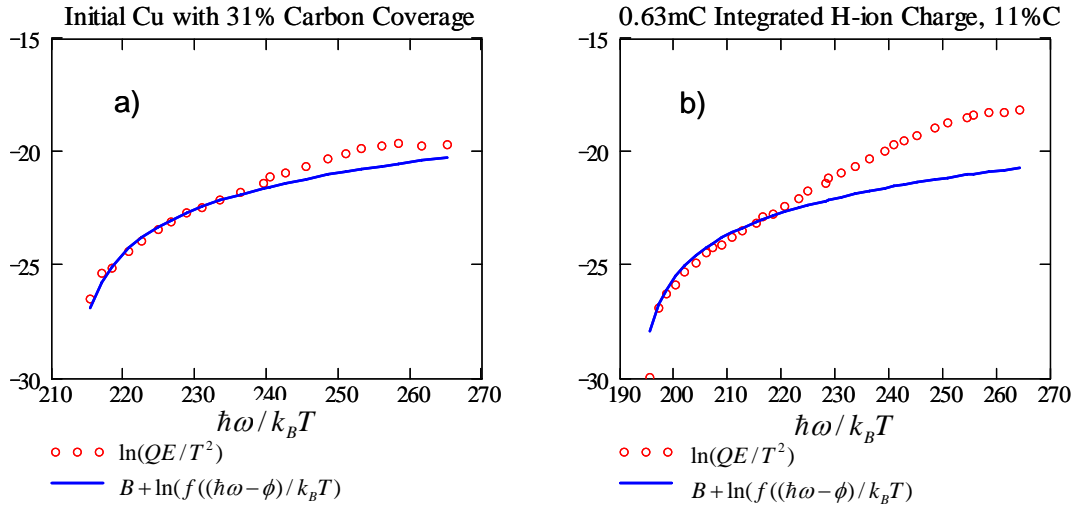


Figure 2: Fowler plots for: a) the initial sample with 31% carbon surface coverage, b) the same sample after exposure to 0.63mC of H-ion beam.

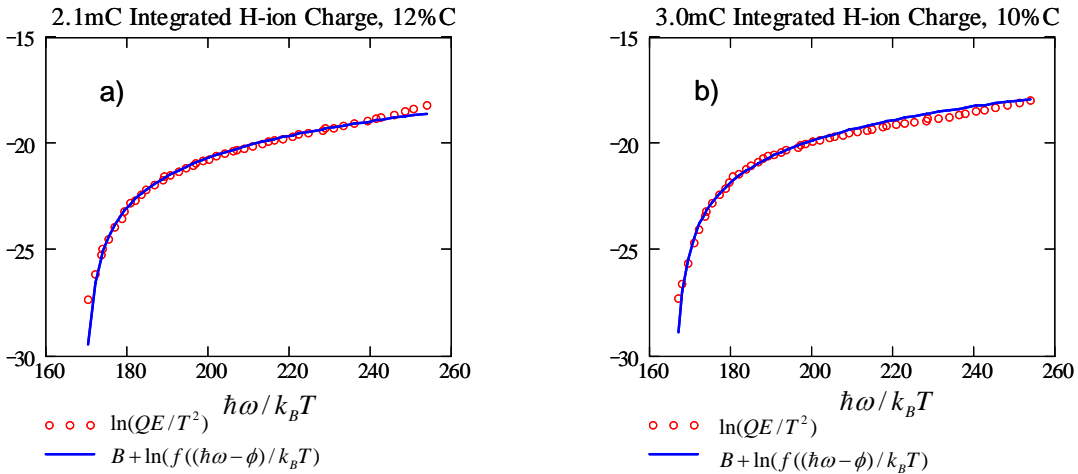


Figure 3: Fowler plots for sample exposed to a) 2.1mC of H-ions and b) after 3.0mC of H-ions.

Another discrepancy is seen at low photon energies in Figure 4b after 10.23mC of H-ions and then heating the sample to 230degrees C for 1hour. The bake was expected to further improve the QE, but instead the QE went down and the work function increased 0.18eV. The Fowler plot indicates a poor fit at the lowest photon energies. Unlike the analysis in Figure 2, it seemed inappropriate to bias the fit to the low energies since this would grossly under-estimate the bulk of the data at higher energies. We speculate this bake re-contaminated the cathode either from out gassing of the surrounding adjacent chamber walls (only the sample was heated to 230degreesC) or by diffusion of material from the sample's interior. However, it should be noted that previous measurements showed heating a contaminated sample to 230degreesC in a clean UHV system resulted in QE's similar to those obtained with H-ion cleaning [1].

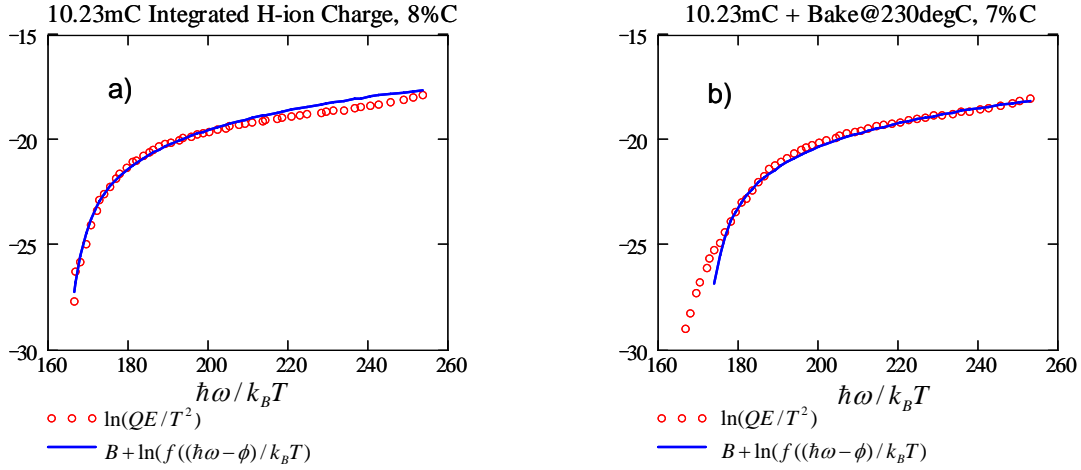


Figure 4: a) Fowler plot after final integrated exposure of 10.23mCoul. b) The result of heating the sample to 230degC.

The work functions obtained for the copper sample as it was progressively cleaned by the H-ion beam are shown in Figure 5. The final work function was 4.31eV with an estimated systematic uncertainty of 0.2eV. There was an increase of 0.18eV when the sample was baked at 230degreesC as described above.

Comparison with an accepted work function for polycrystalline copper is difficult because of the large range quoted in the literature. Therefore an average work function was computed with a standard deviation based upon the reported values [5,6,7,8]. The resulting work function is 4.66+/-0.51eV. Thus our value of 4.31eV is in reasonable agreement.

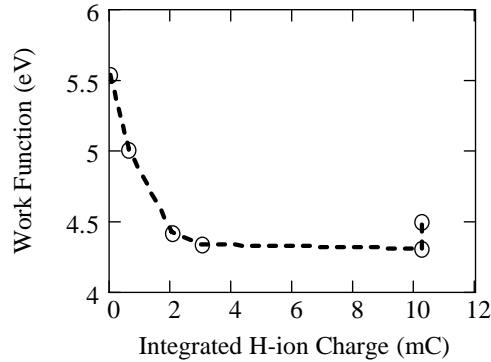


Figure 5: The copper sample work function vs. the accumulated H-ion charge. The higher work function point at 10.23mC was measured after baking the H-ion cleaned sample to 230degC.

DERIVATION OF THE QUANTUM EFFICIENCY FOR A METAL

In previous work [9], the QE was computed using the free electron gas model of a metal and the Fermi-Dirac distribution for the electron density of states (EDOS) [10]. This derivation of QE assumes the electrons are at zero temperature and imposes the requirement that the escaping electron's momentum perpendicular to the surface, p_{\perp} , satisfy [11]

$$\frac{p_{\perp}^2}{2m} > E_F + \phi - \phi_{schottky} \quad [4]$$

where ϕ is the work function, E_F the Fermi energy, and $\phi_{schottky}$ is the barrier energy shift due to the Schottky effect which is conveniently expressed as [12], $\phi_{schottky} = 3.7947 \times 10^{-5} \sqrt{E(V/m)}$ eV. For use in the following discussion, we define the effective work function, $\phi_{eff} = \phi - \phi_{schottky}$.

Our calculation of the QE is based upon the three-step model for photo-emission [13]. In this model, the electron is emitted by means of three sequentially independent processes: 1) absorption of the photon with energy $\hbar\omega$, 2) migration including e-e scattering to the surface, and 3) escape for electrons with kinematics above the barrier. Therefore the QE can be expressed in terms of the probabilities for these steps to occur,

$$QE(\omega) = (1 - R(\omega)) \frac{\int_{E_F + \phi_{eff} - \hbar\omega}^{\infty} dE N(E + \hbar\omega)(1 - f_{FD}(E + \hbar\omega))N(E)f_{FD}(E) \int_{\cos \theta_{max}(E)}^1 d(\cos \theta) F_{e-e}(E, \omega, \theta) \int_0^{2\pi} d\Phi}{\int_{E_F - \hbar\omega}^{\infty} dE N(E + \hbar\omega)(1 - f_{FD}(E + \hbar\omega))N(E)f_{FD}(E) \int_{-1}^1 d(\cos \theta) \int_0^{2\pi} d\Phi} \quad [5]$$

In the first step, a photon is absorbed by a metal with an optical reflectivity of $R(\omega)$ with a probability of $(1-R(\omega))$. F_{e-e} is the probability that an electron reaches the surface without scattering with another electron. In the near-threshold regime we are concerned with, an electron-electron scattering event is not likely to result in either participating electron retaining energy sufficient to escape the material. And third is the probability that an electron will be excited by the photon into a state with sufficient perpendicular momentum to escape the material. Here we follow the method of Bergund and Spicer [10] in assuming that the probability of excitation of an electron with initial energy E to final energy $E+\hbar\omega$ is solely a function of the number of filled states at energy E , $N(E)f_{FD}(E)$, and the number of empty states at energy $E+\hbar\omega$, $N(E+\hbar\omega)(1-f_{FD}(E+\hbar\omega))$. Here $f_{FD}(E)$ refers to the Fermi-Dirac distribution function,

$$f_{FD}(E) = \frac{1}{1 + e^{(E-E_F)/k_B T}} \quad [6]$$

$N(E)$ refers to the number of electron states at energy E , often referred to as the electronic density of states (EDOS). Implicit in this treatment is the assumption that there are no strong selection rules on the momentum states of the electron and that the quantum mechanical matrix element connecting the initial and final electron states is not itself a function of the electron energy. The maximum angle, $\theta_{max}(E)$, at which an electron can approach the surface and still escape is determined by equation 4. The denominator in equation 5 is a normalizing factor that stems from the assumption that every photon that is not reflected leads to an excited electron.

Given that our goal is understanding near-threshold emission from copper, several assumptions can be made to simplify equation 5. First, the EDOS for copper is relatively flat from $\sim 2\text{eV}$ below the Fermi level to the vacuum level [12]. This allows us to assume $N(E)$ and $N(E+\hbar\omega)$ are constant. We assume that the Fermi-Dirac distribution can be taken at $T=0$, resulting in step-like distribution with filled states up to E_F and empty states above E_F . For the near-threshold case considered here, $\theta_{max}(E)$ will be near normal. This allows us to ignore the dependence of F_{e-e} on the exact electron trajectory, and consider it a function of only the photon absorption depth and the electron mean free path for a given photon energy $\hbar\omega$. Below we further simplify the scattering term by taking an average mean free path for the electrons generated by a given photon energy. With these assumptions Eqn. [5] can be separated into three factors corresponding to the three step process,

$$QE(\omega) = (1 - R(\omega)) F_{e-e}(\omega) \frac{\int_{E_F + \phi_{eff} - \hbar\omega}^{E_F} dE \int_{\sqrt{\frac{E_F + \phi_{eff}}{E + \hbar\omega}}}^1 d(\cos \theta) \int_0^{2\pi} d\Phi}{\int_{E_F - \hbar\omega}^{E_F} dE \int_{-1}^1 d(\cos \theta) \int_0^{2\pi} d\Phi} \quad [7]$$

These integrals are easily performed to give [9],

$$QE(\omega) = (1 - R(\omega)) F_{e-e}(\omega) \frac{(E_F + \hbar\omega)}{2\hbar\omega} \left[1 + \frac{E_F + \phi_{eff}}{E_F + \hbar\omega} - 2\sqrt{\frac{E_F + \phi_{eff}}{E_F + \hbar\omega}} \right]. \quad [8]$$

The electron-electron scattering term, F_{e-e} , is derived using the distribution of excited electrons and the electron mean free path due to e-e scattering, after the photon is absorbed into the metal to an optical skin depth, λ_{opt} . Once the photon is absorbed, the excited electrons drift to the metal's surface with some scattering against the valence electrons and losing enough energy to remain bound. The e-e scattering length, λ_{e-e} , or electron mean free path between collisions

with valence electrons determines how many excited electrons reach the surface without any collision. We assume that any collision eliminates the electron's chance to escape. Therefore, the fraction of electrons per unit distance which avoid scattering at a distance s from the surface is given by,

$$f(s) = \frac{1}{\lambda_{opt}} e^{-s \left(\frac{1}{\lambda_{opt}} + \frac{1}{\lambda_{e-e}} \right)}. \quad [9]$$

Integrating into the metal several skin depths gives the fraction of electrons reaching the surface,

$$F_{e-e} = \int_0^{\infty} f(s) ds = \frac{1}{1 + \frac{\lambda_{opt}}{\lambda_{e-e}}} \quad [10]$$

And the expression for the QE then becomes,

$$QE(\omega) = \frac{1 - R(\omega)}{1 + \frac{\lambda_{opt}}{\lambda_{e-e}}} \frac{E_F + \hbar\omega}{2\hbar\omega} \left[1 + \frac{E_F + \phi_{eff}}{E_F + \hbar\omega} - 2 \sqrt{\frac{E_F + \phi_{eff}}{E_F + \hbar\omega}} \right]. \quad [11]$$

The optical skin depth depends upon wavelength and is given by, $\lambda_{opt} = \frac{\lambda}{4\pi k}$, where k is the imaginary part of the complex index of refraction, $\eta = n + ik$ and λ is the free space photon wavelength.

The electron-electron scattering length is strongly energy dependent and can be derived from the Pauli exclusion principle, which constrains the final state energies of both the valence and excited electrons to above the Fermi level. Here we solve for the energy dependence of the e-e scattering length assuming the excited electron scatters only with valence electrons (electrons below the Fermi level) and any scattering event eliminates the electron from escaping. The probability for e-e scattering of an electron with energy E is proportional to the number of occupied initial and unoccupied final electron states for a valence electron with energy E_0 , as well as the availability of vacant states at $E_0 + \Delta E$ and $E - \Delta E$, for the final valence and excited electron states,

$$P(E, E_0, \Delta E) \propto N(E_0) f_{FD}(E_0) N(E_0 + \Delta E) (1 - f_{FD}(E_0 + \Delta E)) N(E - \Delta E) (1 - f_{FD}(E - \Delta E)). \quad [12]$$

As above, we assume that the EDOS is constant, and evaluate the Fermi-Dirac functions at $T=0$. The probability can then be written in terms of Heaviside-step functions, $H(x)$ [14],

$$P(E, E_0, \Delta E) \propto H(E_F - E_0) H(E_0 + \Delta E - E_F) H(E - E_0 - 2\Delta E). \quad [13]$$

The first factor on the right expresses that the valence electron's initial energy must be less than the Fermi energy, $E_0 < E_F$, the second step function requires that the energy gained by the valence electron from scattering with the excited electron must increase the valence electron's energy over the Fermi level, $E_0 + \Delta E > E_F$, and the third factor only allows scattering of the valence electrons to unoccupied states with energies less than the excited electrons final energy level, $E - E_0 < 2\Delta E$. This prevents double counting cases where the valence electron ends up with higher energy than the excited electron due to the indistinguishable nature of electrons. Therefore the energy exchanged is bounded, $E_F - E_0 < \Delta E < (E - E_0)/2$ and the initial valence electron energy constrained to, $2E_F - E < E_0 < E_F$.

The total probability an electron with energy E scatters is then given by,

$$P(E) = \int dE_0 \int d(\Delta E) P(E, E_0, \Delta E) \propto \int_{2E_F - E}^{E_F} dE_0 \int_{E_F - E_0}^{(E - E_0)/2} d(\Delta E) \quad [14]$$

Performing these trivial integrals gives,

$$P(E) \propto \frac{1}{4} (E - E_F)^2 \quad [15]$$

The electron scattering length, λ_{e-e} , is found by using Fermi's Golden Rule [15] which relates the transition rate, $\tau(E)$, to the electron velocity, $v(E)$, and the inverse of the transition probability, $P(E)$, [12]

$$\lambda_{e-e}(E') = v(E')\tau(E') \propto \frac{v(E')}{P(E')} \propto \frac{1}{E'^{3/2}}. \quad [16]$$

Following the discussion and derivation given in Ref 16, we assume the energy for the electron in the conduction band is relative to the Fermi level and compute its velocity, $v(E-E_F) = \sqrt{2(E-E_F)/m}$, and define $E' = E - E_F$. Normalizing the scattering length to a known value at energy E_m above the Fermi level gives the energy dependent electron scattering length,

$$\lambda_{e-e}(E') = \lambda_{e-e}(E_m) \left(\frac{E_m}{E'} \right)^{3/2}. \quad [17]$$

This result is plotted in Figure 6, illustrating the energy dependence of the e-e scattering length. This expression for the e-e scattering length is only valid for electron energies near the Fermi level. Above $E' \sim 50\text{eV}$ the scattering length increases with energy [17, 18].

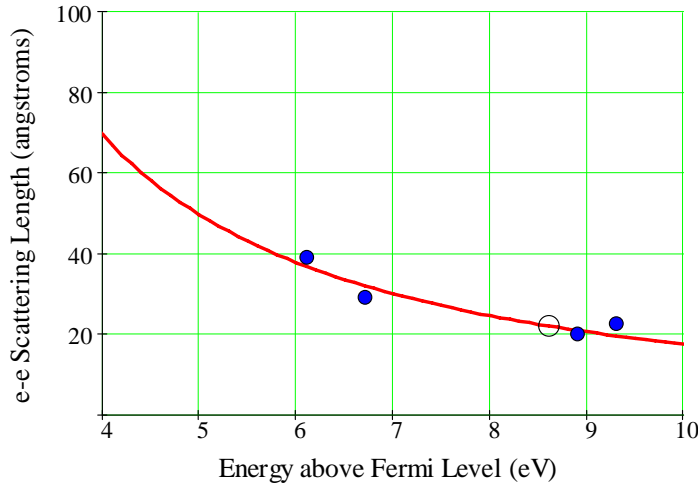


Figure 6. The electron-electron scattering length for copper, plotted as a function of electron energy above the Fermi level in the energy range of interest. The length is computed using a Fermi gas model at $T=0$ as given by Eqn. 16 which has been normalized to $\lambda_{e-e}(E_m = 8.6\text{eV}) = 22 \text{ angstroms}$ indicated by the open circle [16]. Measurements in this energy range are plotted as solid circles [19].

Since the excited electrons have energies between ϕ_{eff} and $\hbar\omega$ relative to the Fermi level, we evaluate F_{e-e} at the average electron-electron scattering length,

$$\bar{\lambda}_{e-e}(\hbar\omega) = \frac{\int_{\phi_{eff}}^{\hbar\omega} \lambda_{e-e}(E) dE}{\int_{\phi_{eff}}^{\hbar\omega} dE} = \frac{2\lambda_m E_m^{3/2}}{\hbar\omega \sqrt{\phi_{eff}}} \frac{1}{\left(1 + \sqrt{\frac{\phi_{eff}}{\hbar\omega}}\right)} \quad [18]$$

Thus our final expression for the QE becomes,

$$QE(\omega) = \frac{1 - R(\omega)}{1 + \frac{\lambda_{opt}}{2\lambda_{e-e}(E_m)} \frac{\hbar\omega \sqrt{\phi_{eff}}}{E_m^{3/2}} \left(1 + \sqrt{\frac{\phi_{eff}}{\hbar\omega}}\right)} \frac{(E_F + \hbar\omega)}{2\hbar\omega} \left[1 + \frac{E_F + \phi_{eff}}{E_F + \hbar\omega} - 2\sqrt{\frac{E_F + \phi_{eff}}{E_F + \hbar\omega}}\right]. \quad [19]$$

COMPARISON OF THEORY WITH EXPERIMENT

The comparison of theory with the experimental results for the 10.32mC H-ion data is given in Figure 7. The theory QE curves are plotted for zero applied electric field, $\phi_{schottky}=0$, corresponding to the field the QE measurements were performed at, and for 50MV/m as is usually obtained in an RF gun operating with a peak electric field of 100MV/m and a launch phase of 30degrees relative to the zero crossing RF phase. For 50MV/m, $\phi_{schottky}=0.268\text{eV}$. In this calculation the optical depth and the optical reflectivity are determined from the wavelength-dependent complex index of refraction [20] in Eqn. 19 to obtain the theoretical curves shown. The agreement is excellent for the entire wavelength range. Therefore, we conclude the above described theory is reasonably accurate and the H-ion cleaning technique produces a nearly ideal, atomically clean copper surface.

Table I. Parameters used to compute the theory curves in Figure 7.

Fermi Energy	7eV
Work Function	4.31eV
$\phi_{schottky}$ @50MV/m	0.268eV
e-e scattering length @8.6eV	22 angstroms

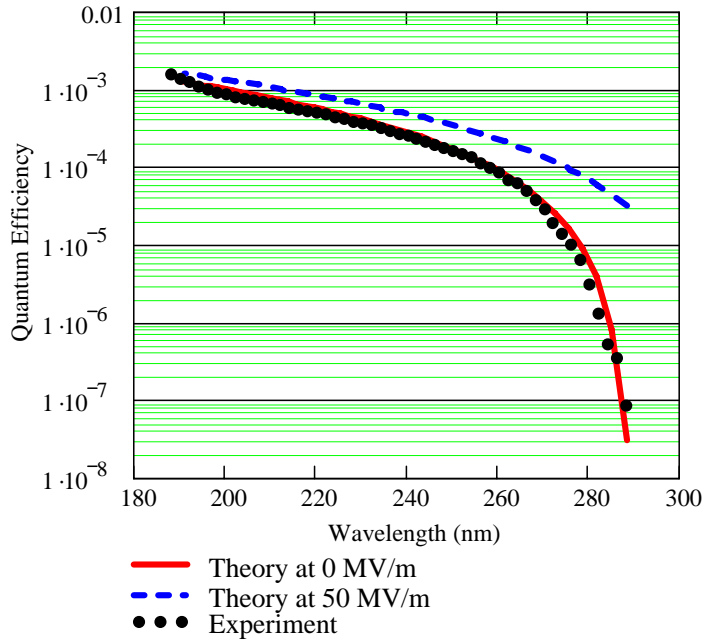


Figure 7: Plot comparing the measured QE at low field (points) and computed QE's using Eqn. [19] and the parameters listed in Table I at low (red) and high (blue) applied fields.

IMPLIMENTING THE H-ION CLEANER ON A S-BAND RF GUN

Figure 8 shows a possible method for implementing an H-ion gun on existing 1.6cell, s-band (2.856GHz) guns. The H-ion gun working distance is limited to approximately 15cm, thus transporting the beam through the solenoid is not possible. However, since nearly all such guns are built with grazing-incidence laser ports on the cathode cell, we propose using one of these ports for directing the H-ion beam onto the cathode. The vacuum pressure increases to the 10^{-4} Torr range during H-ion beam operation and thus, a valve (not shown) is required downstream of the gun and solenoid to protect the rest of the beamline. Work is in progress to investigate installing this type of cleaner on the Linac Coherent Light Source (LCLS) gun. Further details of the LCLS gun can be found in Ref. 21.

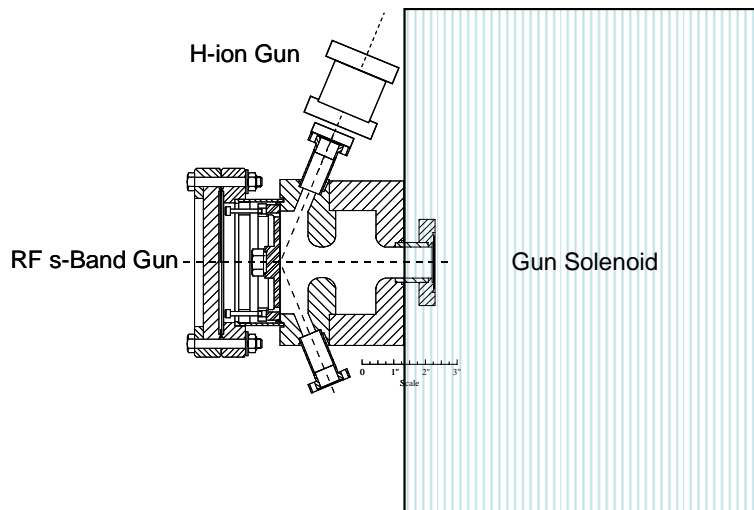


Figure 8: The proposed configuration for in-situ cleaning of the cathode in an s-band RF gun. An H-ion gun with a valve and extension vacuum spool is shown attached to one of the grazing incidence laser ports on the cathode cell.

SUMMARY AND CONCLUSIONS

This work demonstrates the potential of using a 1keV hydrogen ion beam to clean the surface of metal cathodes. Measurements of the QE as a function of optical wavelength were performed as a function of the integrated exposures of a copper sample to the hydrogen beam. The QE reached its maximum value after an exposure of 10.32mC in an area approximately 1cm in diameter. Analysis of the data yielded a work function of 4.31eV, slightly lower than but within the experimental uncertainty of the published work function for copper.

In order to understand the fundamental contributions to the QE, a free-electron Fermi gas model was used to derive a relation for the QE. The derivation was based upon the three-step model of photo-emission and produces results in exceptional agreement with the measurements. This is due in part to using the experimentally determined work function and the lack of features in the electron density of states of copper near the Fermi level. The results of these studies show that H-ion beam cleaned metal cathodes perform close to the theoretical expectations of an atomically clean metal surface.

Based upon this work, it is proposed to install an H-ion gun on the standard s-band RF gun for in situ cleaning of the cathode after its installation. A natural location is one of the laser ports located on the cathode cell of most s-band guns. With the H-ion gun permanently mounted in this location, the cathode can be cleaned whenever it becomes contaminated and the QE is unacceptably low.

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