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Metal Photocathodes for Free Electron Laser Applications

by

Corin Michael Ricardo Greaves

A dissertation submitted in partial satisfaction of the
requirements for the degree of
Doctor of Philosophy

in

Applied Science and Technology

in the

Graduate Division

of the

University of California, Berkeley

Committee in charge:

Professor Roger Falcone, Chair
Professor David Attwood
Professor Ka-Ngo-Leung
Dr. Howard Padmore

Fall 2012

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Corin Michael Ricardo Greaves

Abstract

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University of California, Berkeley

Professor Roger Falcone, Chair

Synchrotron x-ray radiation sources have revolutionized many areas of science from elucidating the atomic structure of proteins to understanding the electronic structure of complex materials such as the cuprate superconductors. These advances have been possible because of the high brightness of synchrotron radiation. This high brightness comes from the very small size and divergence of the electron beam, and the interference of light produced by transverse acceleration of the beam in the periodic magnetic structures in the straight sections of a storage ring. The radiation produced is however a sum over the emission from individual electrons as there is no phase relationship between the positions of each electron; the emission therefore scales as the number of electrons. In a Free Electron Laser (FEL), the main difference to the synchrotron radiation mechanism is that the light field acts on the electron beam, over a long distance in an undulator, and causes electron bunching at the optical wavelength. Electrons in different parts of the electron bunch are therefore correlated, and so emit coherently, with a brightness that scales as the square of the number of electrons. This coherent emission process results in the FEL having a brightness typically 10 orders of magnitude higher than a synchrotron radiation source. Although the FEL concept has been around since the mid 1970's, only within the last few months has the world's first x-ray FEL lased. This FEL, the Linac Coherent Light Source (LCLS) at the Stanford Linear Accelerator Center (SLAC), produces x-rays up to an energy of around 8.5 keV. A much lower energy FEL (FLASH) capable of operation to a few 100 eV started to operate at the DESY laboratory several years ago.

In order to lase, the electron beam in a FEL must have a transverse geometric emittance less than the wavelength of the light to be produced. For the generation of x-ray wavelengths, this is one of the most difficult challenges in the design and construction of a FEL. The geometric emittance can be "compressed" by acceleration to very high energy, but with the penalty of very large physical size and very large cost. The motivation for this work was provided by the desire to investigate the fundamental origin of the emittance of an electron beam as it is born at a photocathode. If this initial, or "thermal" emittance can be reduced, the energy, scale and cost of accelerators potentially would be reduced. As the LCLS used

copper as its photocathode, this material was the one studied in this work. Copper was used in the LCLS as it represented a "robust" material that could stand the very high accelerating gradients used in the photoinjector of the FEL. Metals are also prompt photoemitters, and so can be used to produce very short electron bunches. This can be a useful property for creation of extremely short FEL pulses, and also for creation of beams that are allowed to expand under space charge forces, but in a way that results in linear fields, allowing subsequent recompression.

An ideal photocathode for FEL photoinjector should have high quantum efficiency (QE), small emittance, fast temporal response, long lifetime, and minimal complexity. High QE of cathodes require less power for driving laser and also reduce the risk of damaging the cathode materials. Small emittance reduce the scale of the accelerator, therefore, the cost. Metal photocathodes such as copper exhibit long lifetime and fast response, but have quite low quantum efficiency ($< 10^{-4}$). The aim in this work was to understand the quantum yield of the metal, and the transverse momentum spectrum, as the product of the latter and the cathode beam spot size gives the transverse emittance. Initial x-ray diffraction work provided evidence that the LCLS photocathode consisted of large low index single crystal grains, and so work focused on the study of single crystals that could be produced with atomically ordered surfaces, rather than a polycrystalline material. Present theories of quantum yield and transverse emittance assume the basic premise that the metal is entirely disordered, and work here shows that this is fundamentally incorrect, and that the order of the surface plays a critical role in determining the characteristics of emission. In order to investigate these surfaces, I constructed a laser - based ultra-low energy angle resolved photoemission system, capable of measuring the momentum spectrum of the emission and wavelength and angle dependent electron yield. This system has been commissioned, and data taken on low index surfaces of copper.

Results from this work on single crystal copper demonstrates that emitted electrons from the band structure of a material can exhibit small emittance and high quantum efficiency. We show that the emission from the Cu(111) surface state is highly correlated between angle of incidence and excitation energy. This manifests itself in the form of a truncated emission cone, rather than the isotropic emission predicted from the normal model. This clearly then reduces the emittance from the normal values. It also results in extremely strong polarization dependence, with p-s asymmetry of up to 16 at low photon energy. It also directly suggests ways through changing materials, or by material design to significantly reduce emittance, at the same time increasing electron yield. These results show the benefits that could be gained from electronic engineering of cathodes and should have direct impact in the design of future FEL photoinjectors.

To
Mom and Dad

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Chapter 1

The Free Electron Laser (FEL)

1.1 Introduction

Generation of synchrotron radiation is responsible for much notable advancement in science in the fields of biology, physics and chemistry. Results from synchrotron experiments have investigated atomic structure of proteins and revolutionized the development and characterization of material systems. Such advancements are directly attributable to the brightness achieved by synchrotron radiation sources. The photons produced are the result of accelerating beams of electrons at near-relativistic speeds, through periodic magnetic fields in an instrument called an undulator. The result is a tunable, laser-like beam of photons whose harmonic spectrum is the result of incoherent interference of radiation produced in each period of the undulator. The brightest beams produced by third generation synchrotrons have a brightness of $10^{25} \frac{\text{photons}}{(\text{sec} \cdot \text{mrad}^2 \cdot \text{mm}^2)}$, 0.1% BW) and tunable through wavelengths of a few eV to a few hundreds keV.

The Free Electron Laser (FEL) provides the next frontier of high brightness sources. First proposed in the 1970s, the FEL promises higher brightness through the coherent production of radiation as the electron beam traverses the undulator. As shown in Figure 1.1, this coherence is achieved due to the bunching of the electrons at the photon beam wavelength. The photons co-propagating in the electron beam "act back" on the electrons through the pondermotive potential causing this bunching effect. A "fully bunched" beam corresponds to the saturation of the FEL gain, termed the saturation regime of operation. In this regime, the photons produced in adjacent bunches interfere coherently, yielding an overall peak brightness proportional to N^2 , where N represents the number of electrons present in the electron beam. This represents 10 orders of magnitude greater brightness than is possible from synchrotron radiation.

One drawback of current FELs is the machine scale. The Linac Coherent Light Source (LCLS) at the Stanford Linear Accelerator Center (SLAC), commissioned in 2010, requires more than one kilometer of cascaded electron acceleration and beam conditioning apparatus in order to obtain an electron beam of a quality suitable for FEL lasing. Specifically, an

electron beam parameter called the transverse emittance, $\varepsilon_{electrons}$, must follow the relation:

$$\varepsilon_{electrons} < \frac{\text{FEL wavelength}}{4\pi} \quad (1.1)$$

in order to achieve the maximum beam brightness possible by FELs. The foundation for this requirement is apparent when the phase spaces of the electron and photon beams are compared.

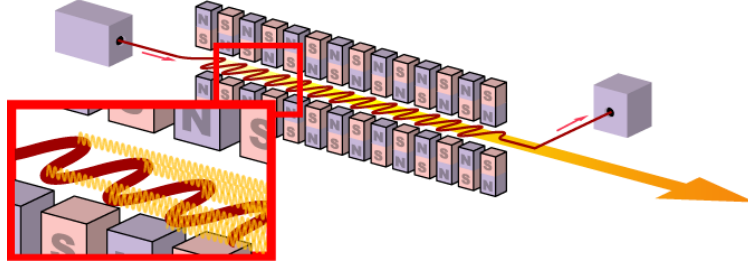


Figure 1.1: A schematic depiction of an Undulator of a Free Electron Laser. Electrons traverse the periodic magnetic fields within the undulator, causing acceleration in the transverse direction. Photons created in the transverse acceleration induce bunching of the electron beam at the photon wavelength through the pondermotive potential. Subsequent oscillations of the bunched beam result in laser-like coherent emission.

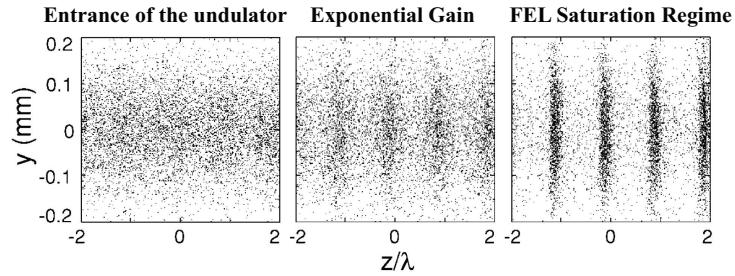


Figure 1.2: Simulation results of electron bunching within the undulator due to the pondermotive potential. The FEL Saturation regime is responsible for coherent emission.[1]

1.2 Emittance and Phase Space

The dynamics of the interactions of electron and photon beams can be characterized by an area distribution in angle-position space called phase space. An area in this 6 dimensional space (angle, position for each spatial dimension), is a complete description of the momenta

of both electrons and photons within the beam. As the beam is modified through focusing elements and acceleration, the aspect ratio of the phase space changes yet its volume remains invariant. The transverse emittance is defined in relation to this phase space area:

$$\varepsilon_{electron} = \frac{\text{Phase space area}}{\pi} = \sigma_x \sigma_x' \quad (1.2)$$

For a photon beam, the emittance is given in the diffraction limit:

$$\varepsilon_{photon} = \frac{\lambda_r}{4\pi} \quad (1.3)$$

where λ_r is the radiation wavelength. Longitudinal acceleration of an electron beam has the effect of compressing the phase space area, consequently reducing the emittance. The change in emittance due to longitudinal acceleration can be approximated in relation to the total beam momentum p_{total} by:

$$\sigma_{x,y}' = \frac{\sigma_{px,y}}{p_{total}} \simeq \frac{\sqrt{\langle p_{x,y}^2 \rangle}}{\gamma mc} \quad (1.4)$$

where the Lorentz parameter $\gamma = \frac{p_{total}}{mc}$, expresses the energy of the near-relativistic beam energies. The Lorentz parameter relates the final beam emittance to the initial emittance (1.2) through the normalized emittance ε_n :

$$\varepsilon_n \simeq \gamma \varepsilon_{electron} \simeq \gamma \sigma_{x,y} \sigma_{x,y}' \quad (1.5)$$

Using (1.4) and (1.5), the normalized emittance can be expressed as:

$$\varepsilon_n = \sigma_{x,y} \frac{\sqrt{\langle p_{x,y}^2 \rangle}}{mc} \quad (1.6)$$

indicating the relationship between normalized emittance and the initial transverse momentum of the electrons within the beam. This fundamental equation summarizes a significant technical challenge in free electron laser design: the normalized emittance, and hence the beam brightness, is inextricably linked to product of the beam size and the initial root mean square momentum of electrons within the beam. Reducing the normalized emittance can be achieved in two ways: reduce the geometric size of the electron beam or reduce the initial root-mean-square momentum distribution of the electrons within the beam. In the language of Equation (1.4), the maximum beam brightness is achieved by the overlap of the electron and photon phase spaces. The impact of this optimization allows the reduction in longitudinal acceleration required for FEL lasing, reducing machine scale and cost. Alternatively, the lasing wavelengths of the FEL could be expanded, providing shorter wavelength operation for a fixed accelerator length.

1.3 Space Charge: The Child Langmuir Law

Reduction of the electron beam size in the first term of Equation (1.6) is effective up to the limit in which an accelerating potential is screened by the charges generated within the accelerating gap. This is termed the Space Charge limit and is governed by the Child-Langmuir Law. Figure 1.3 illustrates this condition. For a generating light pulse of length less than the electron flight time ($\frac{d}{c}$), the photogenerated charge within the gap can be approximated as a sheet of charge. In this condition, a simple relationship for the minimum beam radius can be achieved:

$$r = \sqrt{\frac{Qd}{\pi\epsilon_0 V}} \quad (1.7)$$

For a given amount of charge Q , the minimum electron beam radius and hence the minimum emittance is set by the accelerating gradient ($\frac{V}{d}$) of the structure used to accelerate electrons initially to relativistic velocity. The combination of the cathode, laser and acceleration is termed a photo-gun. Further optimization of the normalized emittance would require a reduction in the required charge of the machine or an increase in the accelerating gradient at the photocathode. Gains in beam brightness due to lower emittance are then left to optimization of the second term of Equation (1.6) through reducing the root-mean-square momentum of the photoemitted electrons. Elucidating the nature of this optimization is the subject of this dissertation.

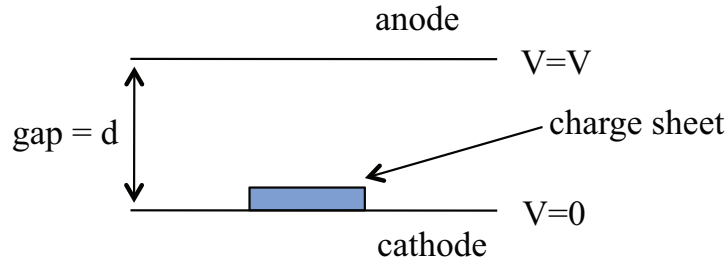


Figure 1.3: A depiction of the condition governing the space charge regime. Screening of the accelerating potential occurs due to the introduction of charges within the accelerating gap.

1.4 Dissertation Outline

The chapters that follow present the theory, experimental setup, data and conclusions drawn from a lab constructed for characterization of photocathodes for FEL applications. Chapter 2 presents the theoretical model of photoemission currently used within the electron beam community to understand the quantum efficiency and angular distribution of

photoemitted electrons. The conclusions of this model are verified by laboratory experiments, the setup and development of which are discussed in Chapter 3 . Chapter 4 shows the methods of analysis used to process data recorded from each experiment. Chapter 5 discusses the significance of the data to current FEL development and presents a path forward for increasing brightness through the engineering of photocathodes. The appendices contain the relevant engineering diagrams, control algorithms and data analysis codes developed in the production of this dissertation.

Chapter 2

The Three Step Model of Photoemission

2.1 The Three Step Model of Photoemission

Spicer Three Step Model of Photoemission is a model commonly used to evaluate candidate metal systems for use as a photocathode. Developed in 1950, the model describes photoemission as a bulk process, relying only on the material work function, electron scattering lengths and dielectric function as variables for fitting experimental data.[2] In practice, this model is successful in predicting relative quantum efficiencies of candidate photoemissive materials and has been extended by Dowell to provide estimates of thermal emittance of the photoemitted electron beams.[3, 4, 5] In this section, the physics of the model is reviewed and its impact on electron yield and transverse emittance through the photon emission angle are introduced.

Figure 2.1 shows a schematic of the Three Step Model of electron photoemission. They are:

1. Promotion of the electron from the conduction band to the valence band through photoelectric absorption.
2. Transport of the electron from the generation site to the metal-vacuum interface.
3. Emission across the metal-vacuum interface.

Step one describes the physics of the interaction of the photon with the photocathode. The Fresnel equations are used to describe the reflectivity coefficient, $R(\theta, \omega)$, as a function of angle of incidence and photon wavelength. The photon absorption coefficient, $A(\theta, \omega)$, is given by [6]:

$$A(\theta, \omega) = 1 - R(\theta, \omega) \quad (2.1)$$

In calculation of the absorption step, only the initial and final energies of the photon are considered; the crystal momentum of the electron in its initial state is not considered.

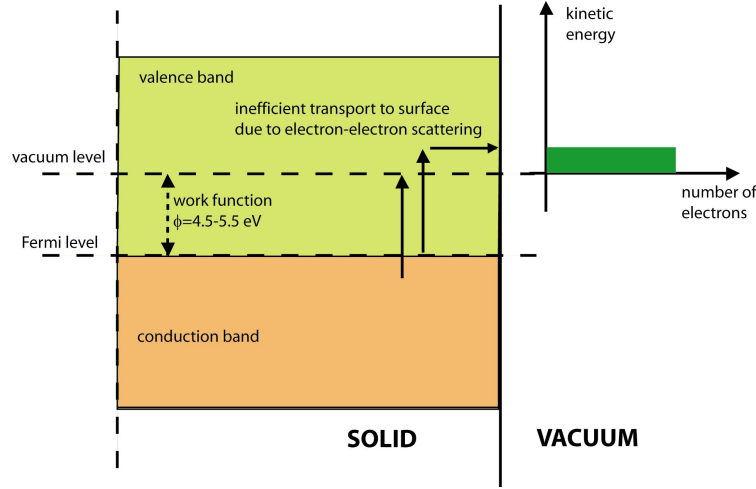


Figure 2.1: A schematic depiction of the three step model of photoemission. Electrons are promoted from the conduction band to the valence band through a photon absorption event. The electron undergoes scattering events as it transits from the production site to the material surface. The electron is then emitted from the surface into the vacuum.

Energy is conserved; momentum is not. Step two describes photon transport to the surface by calculating an electron transit probability, $F_{e-e}(\omega)$. The electron transit probability is a function of both the of the optical absorption length, $\lambda_{opt}(\omega)$, and the dominant scattering cross section λ_{e-e} . Metal systems are dominated by electron-electron scattering in which a single scattering event reduces the electron energy such that escape is no longer possible. The form of the transit probability is therefore:

$$F_{e-e}(\omega) = \frac{1}{1 + \frac{\lambda_{opt}(\omega)}{\lambda_{e-e}}} \quad (2.2)$$

The final step is emission from the bulk into the vacuum. States responsible for photoemission are those whose component of kinetic energy parallel to the surface normal exceeds the surface barrier energy. Integrating over these states and normalizing to all available states results in an emission probability that depends on the square of the excess energy. This is given by:

$$P_{emission} = \frac{(\hbar\omega - \phi_{eff})^2}{8\phi_{eff}(E_F + \phi_{eff})} \quad (2.3)$$

The individual components of the model are evaluated for Copper and Aluminum metal systems up to one volt in excess energy in Figure 2 a-e. The limiting process is the third step and the quantum efficiency therefore depends approximately on the square of the excess energy. The total quantum efficiency is the product of the terms of each steps:

$$QE(\omega) = A \cdot F_{e-e}(\omega) \cdot P_{emission} \quad (2.4)$$

and is shown in Figure 2.2d.

2.2 Predictions of the Three Step Model

The prediction of the Three Step model for the emittance of the produced electron beam is derived using the condition on the perpendicular momentum assumed in step three. Assuming a constant density of states and integrating over available initial and final states and angles as shown:[3]

$$\langle p_x \rangle^2 = \frac{\int_{E_f + \phi_{eff} - \hbar\omega}^{E_f} dE \int_{\cos(\theta_{max})}^1 d(\cos \theta) \int_0^{2\pi} d\Phi p_x^2}{\int_{E_f + \phi_{eff} - \hbar\omega}^{E_f} dE \int_{\cos(\theta_{max})}^1 d(\cos \theta) \int_0^{2\pi} d\Phi} \quad (2.5)$$

and substitution into the expression derived for the emittance in Chapter 1 results in:

$$\varepsilon_n = \sigma_{x,y} \sqrt{\frac{\hbar\omega - \phi}{3mc^2}} \quad (2.6)$$

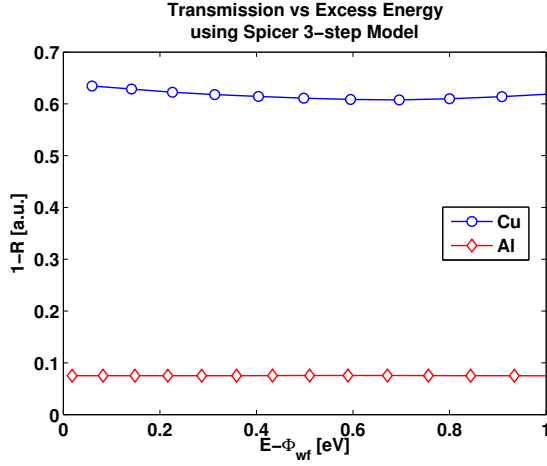
According to the model, the normalized emittance scales as the square root of the excess energy. Low emittance therefore results in near-threshold photoemission. Reducing the excess energy, however, reduces the observed quantum efficiency as the square of the reduction. Therefore, lower emittance also requires lower quantum efficiency. Figure 2.3 illustrates another requirement of the model. The restriction on the initial kinetic energy of the electrons within the bulk defines a maximum internal angle for valid photoemission events. Boundary conditions preserve the component of the kinetic energy tangential to the surface normal. As a result, those electrons traveling at the maximum internal angle emerge traveling parallel to the photocathode surface. *The Three Step model therefore predicts that the momentum distribution of photoemitted electrons outside the cathode is continuous from normal to grazing emission.*

Returning to step one of the model provides insight of the behavior of photoemission as a function of the angle of incidence of the photon beam, for the case of p polarized light due to the reduction in reflectivity. The Fresnel equations cause an increase of the predicted quantum efficiency as a function of angle of incidence. A 25% increase in quantum efficiency can be gained by adopting a 60 degree angle of incidence of illumination on the cathode. (Figure 2.4)

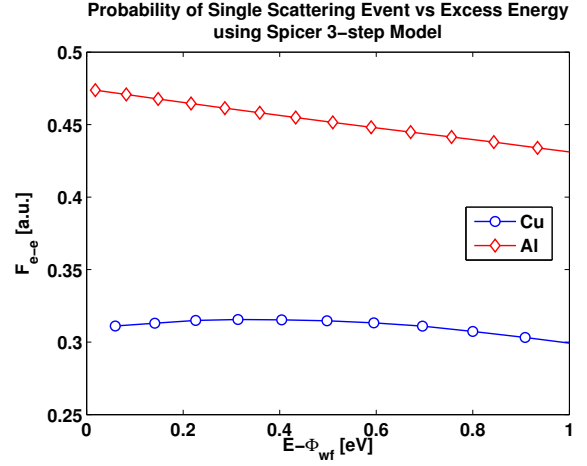
The goal of this work is to verify the behavior of photocathodes as predicted by the Three-Step model, namely that:

1. The momentum distribution of electrons is continuous.
2. The quantum yield of the cathode can be enhanced by varying the angle of incidence of the illumination.

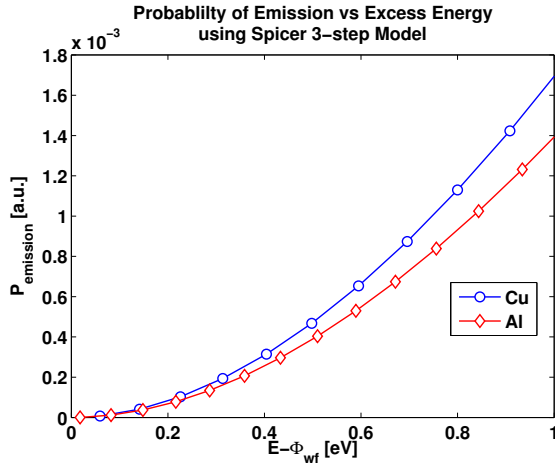
Another aim of this work was to find ways to circumvent the assumptions of the Three-Step model by use of ordered substrates. The implicit assumption in the Three-Step model is that the material is a random array of microscopic domains. In the case of an ordered material one would expect to see manifestations of the bandstructure and an open question investigated here was whether this could be used to advantage. To this end, an experimental laboratory was developed to characterize the quantum efficiency and momentum spectra of photocathode materials. Chapter 3 details the capabilities and instrumentaton of the characterization laboratory.



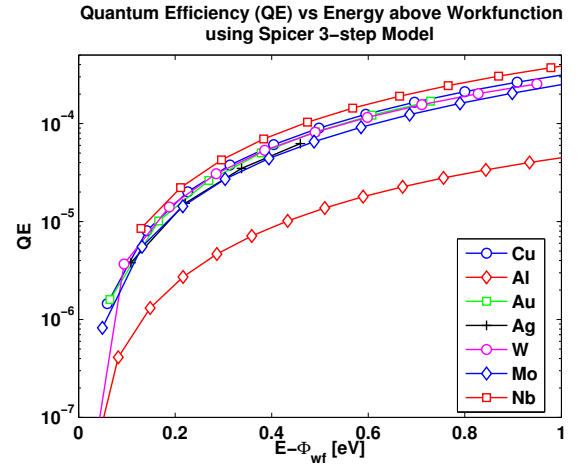
(a) The Transmission coefficient as a function of energy above the work function, evaluated for Copper and Aluminum using the Fresnel Equations. Illumination is assumed to be parallel to the surface normal. (Equation 2.1, $\theta = 0$)



(b) The electron extinction probability due to a single scattering event as a function of energy above the work function, evaluated for Copper and Aluminum using the form of the scattering probability. (Equation 2.2)



(c) The probability of electron emission as a function of photon energy above the work function. This step dominates the behavior of the quantum efficiency. (Equation 2.3)



(d) The Quantum Efficiency for candidate photocathodes predicted by the Three Step Model of Photoemission. (Equation 2.4)

Figure 2.2: Individual steps of the Dowell-Spicer Three Step Model of Photoemission evaluated for candidate photocathode materials.

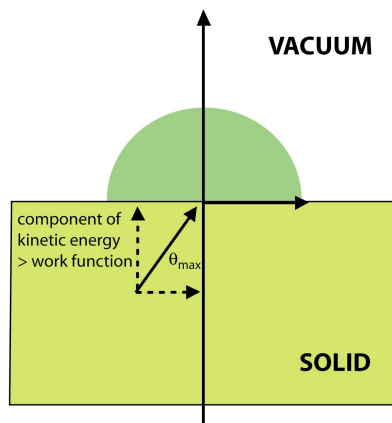


Figure 2.3: The refraction of the electron due to restrictions imposed by boundary conditions at the cathode-vacuum surface. Electrons traveling at the maximum internal angle of escape emerge parallel to the surface, thus defining a maximum internal angle of escape.

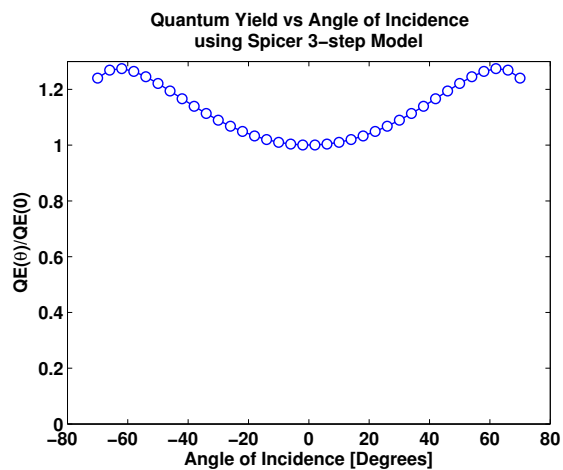


Figure 2.4: The prediction of the total quantum yield of the Dowell-Spicer Three step model of Photoemission, calculated by varying the angle of incidence of illumination as prescribed by the Fresnel Equations for p polarized light. Note the very weak dependence on angle. (0 Degrees = Normal Incidence)

Chapter 3

Experimental Setup

3.1 Introduction

The Photoemission Characterization Laboratory includes sample preparation capabilities and utilizes three major experiments to investigate the assertions of the Three Step Model of Photoemission for metal photocathodes. First, the surface orientation and sample cleanliness are verified using Low Energy Electron Diffraction (LEED) and measurement of the sample work function. Secondly, the Angle Resolved Electron Yield (AREY) experiment is used to investigate the enhancement in quantum efficiency as a function of angle differing from that of the Fresnel Equations. Finally the electron momenta in the photoemitted beam, below the space charge limit, is verified using Time of Flight Angle Resolved Photoelectron Spectroscopy (TOF-ARPES).

Figure 3.1 is a photograph of the characterization laboratory. Six major components comprise the experimental system: the analysis chamber, laser system, harmonic generation optics, Fowler system and control electronics. The experiment layout and optics setup are shown schematically in Figure 3.2. The Ultra High Vacuum (UHV) chamber houses the sample under investigation as well as the sample preparation and analysis instrumentation. The laser plasma source and monochromator provide a tunable CW source of photons (200nm - 1000nm, 3nm bandwidth) for measurement of photocurrent as a function of wavelength. This capability is used in determining the sample work function by the Fowler method as described in Chapter 5. A second laser source provides a pulsed source of tunable photons (800-900 nm) from which the fourth harmonic is derived by two cascaded harmonic generation stages. The pulsed source is required for time of flight measurement of the photoemitted electrons. The system is controlled by a real-time control system.

In this chapter the relevant physics and engineering of the characterization laboratory are presented. First, the experimental chamber and control system are discussed. Discussion of the laser system and relevant physics of the harmonic generation system follows. Next, the details of the work function measurement and ARPES systems are presented. Sample preparation and verification are discussed in the final section of this chapter.

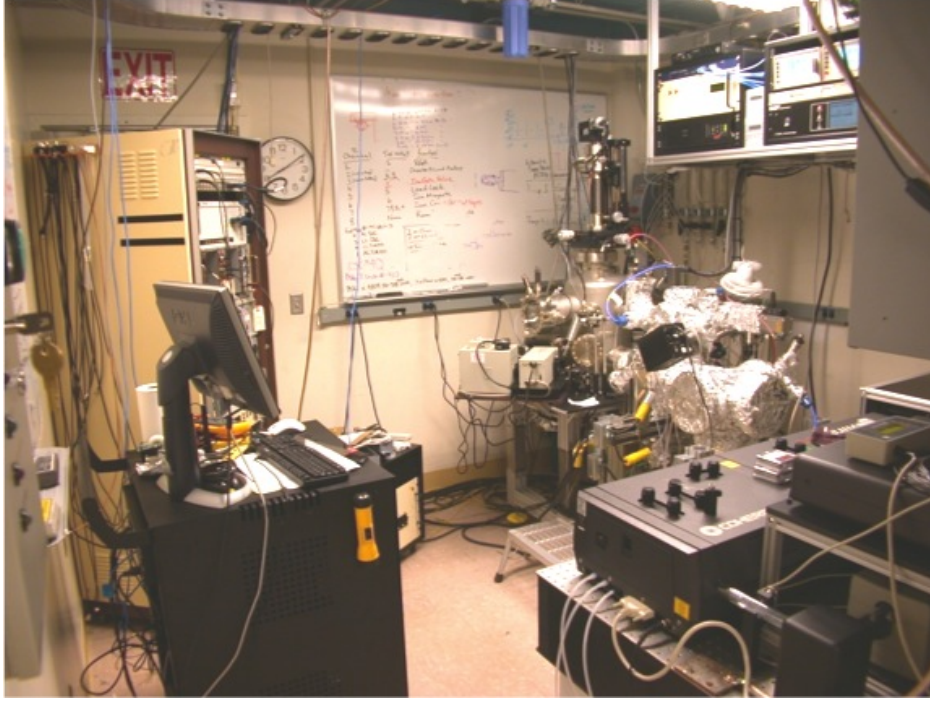


Figure 3.1: A photograph of the Photocathode Characterization Laboratory. The analysis chamber, laser system, control electronics, experiment computer and experiment electronics rack are visible.

3.2 Experimental Chamber and Control System

Characterization of near-bandgap, low kinetic-energy photoemission requires careful attention to both vacuum conditions and residual electric and magnetic fields present in the experimental chamber. For this reason, a custom three-level Mu-Metal UHV chamber was built. Figure 3.3 illustrates the fabricated chamber and labels the position of relevant sample preparation and characterization instrumentation. Samples enter the analysis chamber through the Load Lock Transfer Port on the Preparation Level of the chamber and are captured by an axially mounted 3-axis sample manipulator. From this position, the sample can be translated vertically within the chamber. Ar⁺ ion sputtering, sample annealing and analysis via Low Energy Electron Diffraction (LEED) are also conducted on this level. The second level, the Analysis Level has sapphire viewports (Kurt J, Lesker VPZL-275S) for optical analysis of the sample as well as the Time of Flight detector head. The Pump Level of the chamber houses the Turbo and Ion pumps, non-evaporable getters, roughing valve, nitrogen leak valve and Ion and Pirani gauges. Table 3.1 lists the function and description of the instrumentation used on each level of the Analysis Chamber.

The analysis chamber is capable of reaching base pressures of 2×10^{-10} Torr after a full

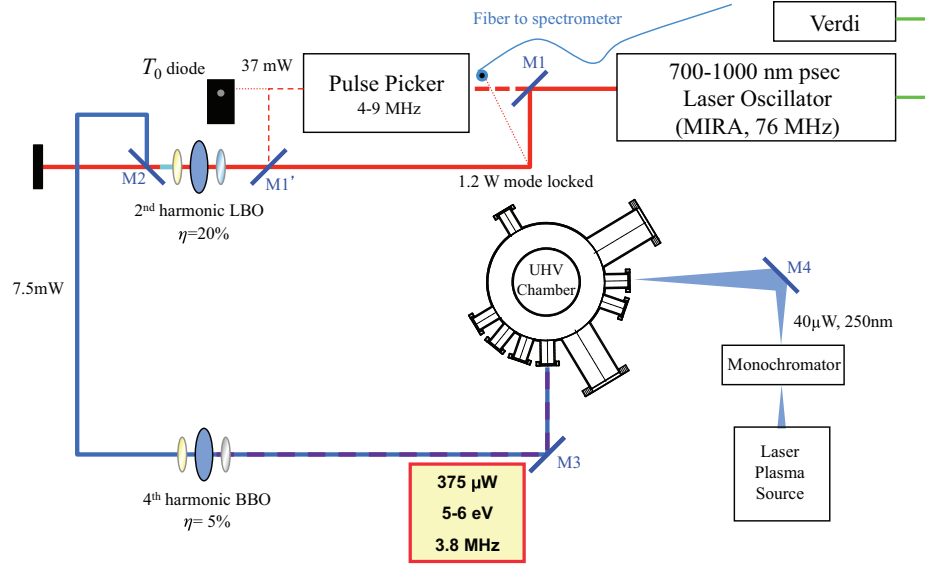


Figure 3.2: Schematic of the layout of the characterization laboratory. A Ti:Sapphire laser pumped by a 10W 532nm Verdi pump laser was used to create a 800-900 nm, 32mW mode locked, 3ps beam. For time-resolved experiments, an acousto-optical pulse picker operating at 3.8MHz was employed. The resulting beam underwent two stages of harmonic generation, resulting in a $375\mu W$ beam at 200-250nm. In addition, a CW laser plasma source focused into a 1:1 monochromator was used to measure sample work function. This system produces $40\mu W$ power at 250nm.

Table 3.1: Instrumentation used in the experiment and the respective chamber locations.

Level	Function	Manufacturer	Model / Description
0	Manipulator	VG	Omniax 3-axis; XL25 Sample Transfer
1	Ion Gun	SPECS GmbH	IQE 11
1	LEED System	SPECS GmbH	ErLEED 150, LaB6 Cathode
2	TOF Detector	Surface Concepts GmbH	DLD 4242
3	Turbopump	Varian Vacuum Systems	Turbo-V 301 Navigator
3	Ion Pump	Gamma Vacuum	
3	Getters	SAES Getters	D-400-2, ST172
3	RGA Detector	Inficon	Transspectre

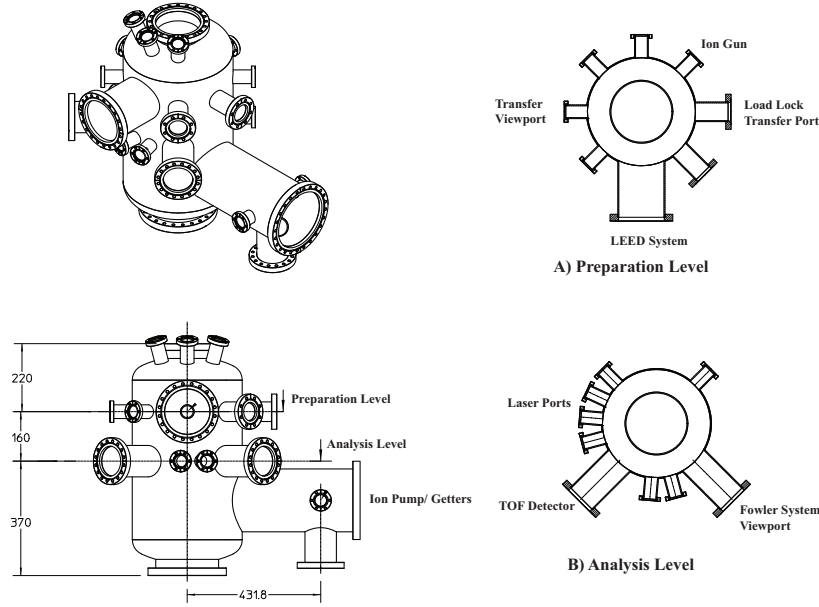


Figure 3.3: The Mu-Metal Chamber fabricated for photocathode characterization. Relevant ports and their respective functions are labeled. Distances are in millimeters. A) shows the port assignment on the preparation level of the chamber. B) illustrates the port assignment on the Analysis level of the chamber.

system bakeout of at 120C for 72 hours. The details of the bakeout system and independent PID controller are discussed in the Appendices. Figure 3.4 shows the LabView control GUI for the experimental system. Figure 3.5 is a picture of the chamber system with 5 dimensional manipulator and vacuum electronics in the upper right. Figure 3.6 shows the sample mounting and temperature control system. Chamber operation, pressure and process monitoring, sample motion and emergency shutdown are controlled by a National Instruments realtime PXI controller and custom built electronics. The realtime controller runs a modified version of the ALS Beamline Control system for motor control and process monitoring. Operation of the chamber components occurs via TCP-IP connection to the experiment computer. Details of the custom hardware and its configuration are listed in the Appendices.

3.3 Laser System

A diagram of the laser system shown in Figure 3.2. A 10 Watt, 532 nm, diode-pumped Coherent Verdi cw-laser pumps a tunable Coherent MIRA Ti:Sapphire oscillator operating in picosecond mode. (1.2W Mode locked power, Central Wavelength 800nm, 76 MHz repetition rate, < 3ps pulse length, s-polarization). Limited by the TOF detector fast electronics, a Coherent Model 9200 acousto-optical pulse picker is used to count down the repetition rate to

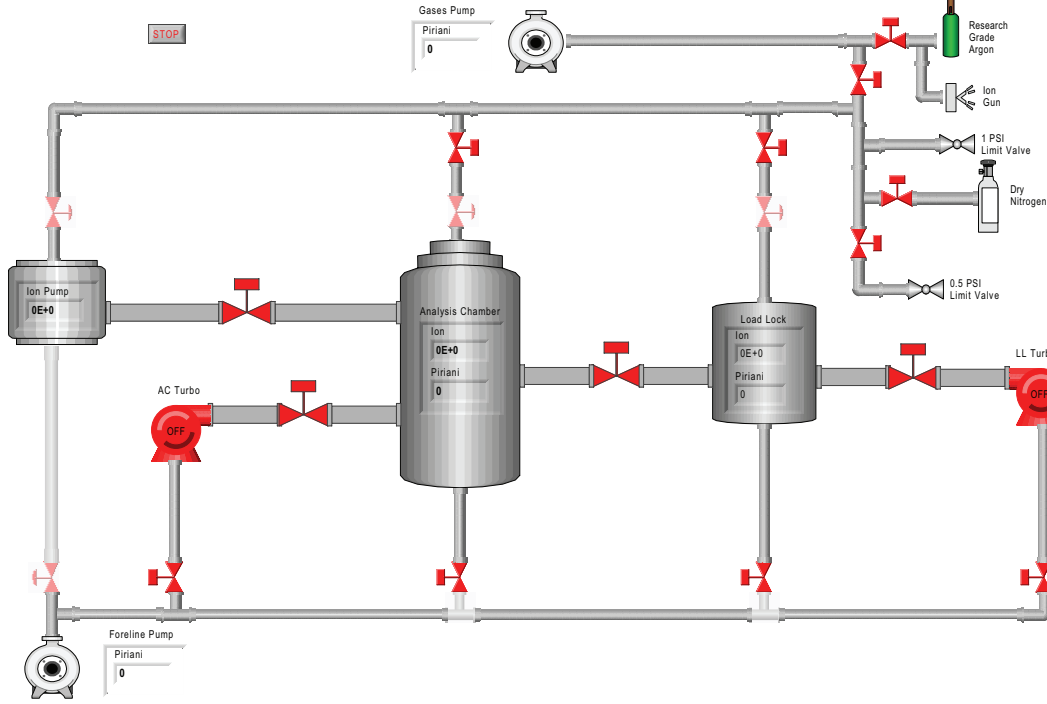


Figure 3.4: A screen capture from the Valve Control HMI.vi Labview control for the characterization system. Pressure readings from each chamber in the system are updated at a refresh frequency of 10Hz. The control valves are actuated by mouse click, and the valve status is represented correspondingly.

a few MHz. The acousto-optic Bragg cell in the pulse-picker provides single-pass diffraction of the pulse train from the oscillator, while the controller allows selection of the repetition rate from 9.75kHz - 4.75 MHz. The oscillator is a modelocked ultrafast laser that uses Ti:Sapphire as the gain medium and is tunable from 700nm - 1000 nm. Figure 3.7 shows the Coherent MIRA laser and frequency doubling and quadrupling setup enclosed in the black box.

3.4 Fourth Harmonic Generation System

Harmonic generation is a process by which harmonics of the wavelength of an incoming photon beam are produced within a nonlinear medium. In such media, the material susceptibility can be expressed as a Taylor expansion:

$$P_{NL}(t) = \chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \dots \quad (3.1)$$

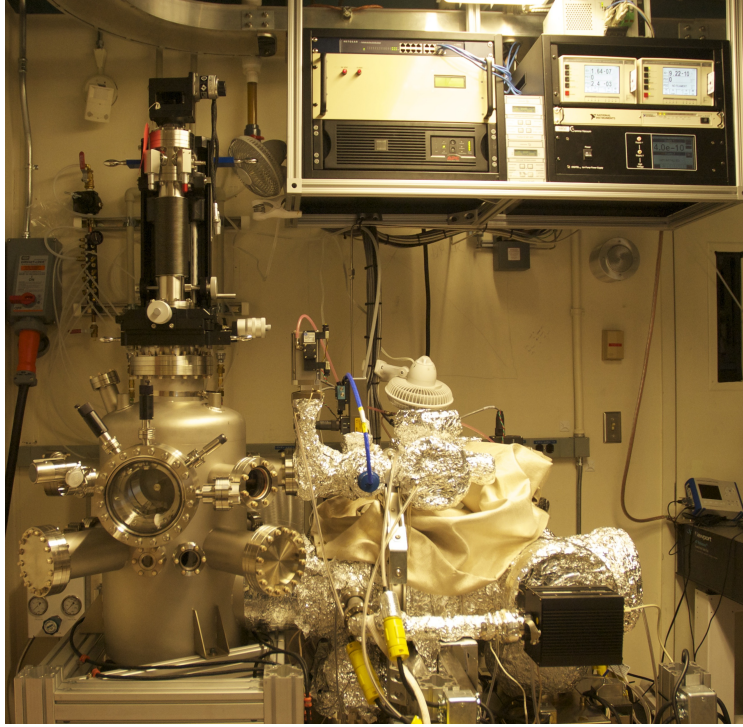


Figure 3.5: The UHV vacuum system and control electronics under construction. The sample sits on a four-axis manipulator in the main chamber and can be inserted and removed using a transfer arm located in the adjacent load lock. The main chamber is pumped by a 300 liter/sec Varian Turbopump, an Ion Pump, and SAES Getters for Hydrogen absorption. Vacuum products are monitored by a residual gas analyzer also shown here.

from which the nonlinear behavior of the material is clearly understood. Here, the coefficients, $\chi^{(n)}$, are the n -th order susceptibilities of the medium. P_{NL} is the dielectric polarization in units of dipole moment per unit volume. The nonlinear solid can be thought of as being comprised of a series of dipole emitters, whose polarization is proportional to the field of the incident (pump) wave. Therefore, over the length scale of the nonlinear crystal, two factors, namely energy density (beam focus) and phase matching of the fundamental and produced harmonics are critical for the efficient production of harmonic beams.[7, 8, 9, 10, 11]

There are two types of angular phase matching methods, depending on the polarization of the input pump beams. Type I phase matching occurs when the pump beams are of the same polarization, resulting in a harmonic beam whose output is polarized in the orthogonal direction (ooe, eeo). Type II phase matching utilizes pump beams of differing polarizations, or a single pump beam whose polarization is at an angle to the optic axis of the nonlinear medium.

In this system, two Type I phased matched Second Harmonic Generation (SHG) crystals

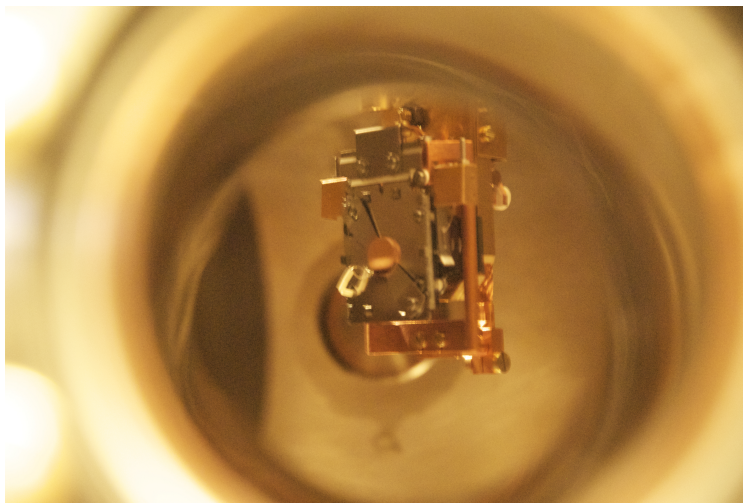


Figure 3.6: A photograph of a Cu(111) sample mounted on the sample chuck and attached to the Manipulator. The sample chuck provides heating and cooling (77 - 1000 K) of the sample, a thermocouple, and a BNC connection for monitoring of the sample photocurrent.

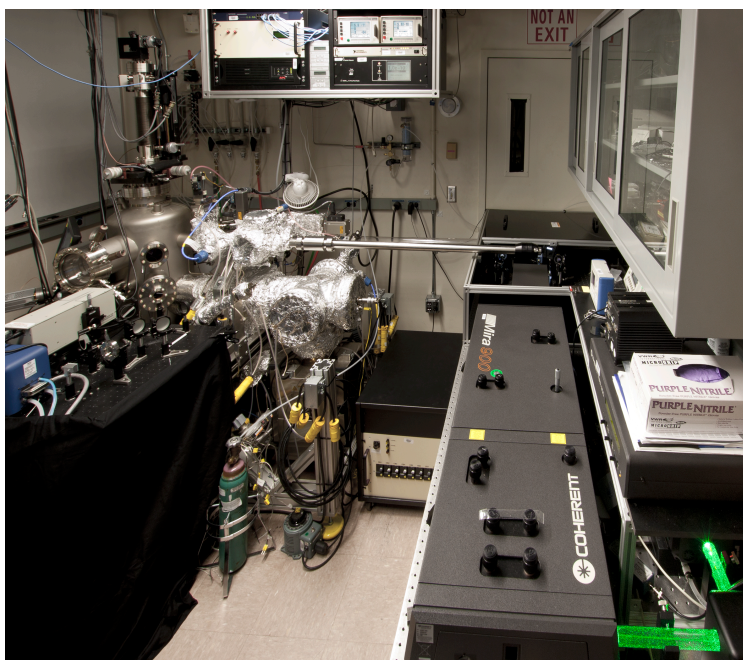


Figure 3.7: The optical bench at the right houses the pump laser, MIRA oscillator and the second harmonic optical setup. The left hand optical table houses the plasma source, monochromator and optics for the work function measurement. Also visible is the system bakeout electronics rack, located between the optical table and the vacuum chamber.

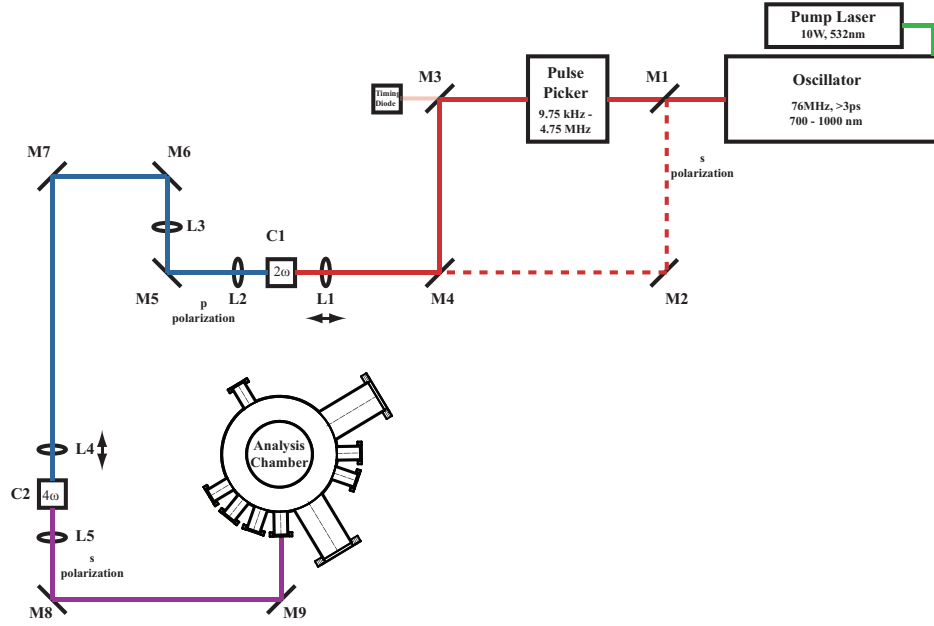


Figure 3.8: A schematic representation of the harmonic generation system. Mirrors and lenses referred to in the text are labeled.

are cascaded to arrive at the fourth harmonic of the fundamental Ti:Sapphire pump beam. For each setup, the crystal cut angles and dimensions have been selected for angular phase matching in the respective wavelength ranges. For the primary conversion (857 to 428 nm), an LBO crystal been employed to preserve beam quality through a reduced walk-off angle, despite its relatively low nonlinear coefficient. For the second stage (428 to 214 nm), a BBO crystal is used. Due to the high sensitivity of the delay-line-detector, very low photon intensity is required, allowing the use of two crystals for our frequency doubling and quadruple setup by ignoring the phase match.

In the harmonic setup, shown in Figure 3.8, the fundamental beam past the pulse-picker and is steered by mirrors M3 and M4, and focused onto the LBO crystal (C1) with lens L1. The combination of lenses L2 and L3 collect the second harmonic beam generated by C1, providing a beam of minimal size and divergence for use in the fourth harmonic setup. Mirrors M5 - M7 are harmonic separators, used to eliminate residual fundamental beam from the second harmonic beam. Lens L4 brings the second harmonic beam to a focus onto Crystal C2, and the resultant fourth harmonic beam is collected by lens L5. The positioning of L5 is also used to affect the resulting size of the beam at the sample. Finally, mirrors M8 and M9 are polarization sensitive harmonic separators, used to separate the residual second harmonic beam from that of the fourth. The resulting beam fourth harmonic beam is then focused onto the sample. Spectral verification of the fundamental, second and fourth harmonic beams are shown as measured by the Ocean Optics Spectrometer, in Figure 3.11.

The parameters of the optics used in the setup are shown in Tables 3.2, 3.3 and 3.4.

Conversion efficiency in each of the harmonic stages is of critical importance. For the second harmonic generation stage, the tuning angle is varied until a maximum power output in the second harmonic is observed, indicating that the phase matching condition is satisfied. Next, the pulse length of the laser is maximized by adjusting the PZT bias of the cavity endmirror. Finally, the energy density within the crystal was maximized by moving the L1 lens and further maximizing the output power. Illustrated in Figure 3.9, the L1 to C1 distance was optimized to within the depth of focus of lens L1, suggesting that the maximum achievable pumping of the second order nonlinear response within C1 is achieved. It can be seen that the maximum conversion efficiency for second harmonic does not change within 1.8mm at the focal distance of L1. The observed conversion efficiency of the second harmonic generation process with the fundamental pump power is shown in Figure 3.10. The input fundamental beam was attenuated using a series of Neutral Density filters, and the power in the second harmonic was monitored after mirror M7. The curve fits well to the square of the power as derived from 3.1.

Table 3.2: Parameters of the Harmonic System Lenses

	Part Number (Thorlabs)	Material	Coating	f [mm]
L1	LB1471-B	BK7	AR 650 1050 nm	50.0 mm
L2	LB1471-A	BK7	AR 350 650 nm	50.0 mm
L3	LB4374	Fused Silica	Uncoated	1000.00 mm
L4	LB1471-B	BK7	AR 350 650 nm	50.0 mm
L5	LB4096	Fused Silica	Uncoated	50.0 mm

Table 3.3: Parameters of the Harmonic Crystals (Ekspla)

	Material	Dimensions	θ	ϕ
C1	LBO	5 mm x 5 mm x 10 mm	90	29
C2	BBO	5 mm x 5 mm x 5 mm	81.5	90

Lens L2 is of critical importance for achieving a proper beam size, shape, and energy density on crystal C2. Because the C1 to L2 distance was constrained, lens L3 was introduced at a distance of 165mm from L2. The combination of L1 and L2 gives the flexibility to achieve the best focus on C2. The equivalent lens focus f_{eff} is given by:

$$\frac{1}{f_{eff}} = \frac{1}{f_1} + \frac{1}{f_2} - \frac{d}{f_1 f_2} \quad (3.2)$$

Table 3.4: Parameters of the Mirrors

	Part Number	Specification	Batch No.
M1	Layertec / 105724	HR (45 700 1010) Turning Mirror	R020812
M2	Layertec / 105724	HR (45 700 1010) Turning Mirror	R020812
M3	Layertec / 105724	HR (45 700 1010) Turning Mirror	R020812
M4	Layertec / 105724	HR (45 700 1010) Turning Mirror	R020812
M5	Layertec / 103948	HR p(45 370 - 425 nm) Separator	R020821
M6	Layertec / 103948	HR p(45 370 - 425 nm) Separator	R020821
M7	Layertec / 103948	HR p(45 370 - 425 nm) Separator	R020821
M8	Newport / BSR-01-1025	Polarizing Beamsplitter	SO#295829 LOT# 470053
M9	Newport / BSR-01-1025	Polarizing Beamsplitter	SO#295829 LOT# 470053

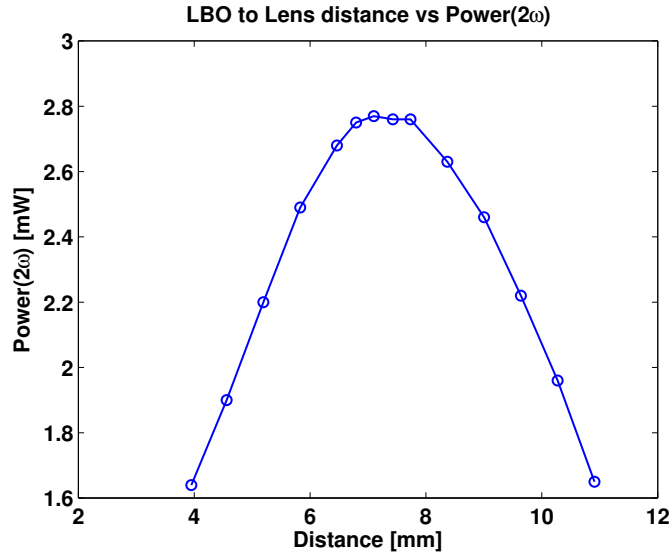


Figure 3.9: Depth of focus of the primary lens within the second harmonic crystal. The lens-to-LBO distance was varied and the power output power was measured, with a fundamental power of 5.3mW. The flattening of the expected parabolic shape indicates the region over which the pump beam remains within critical focus.

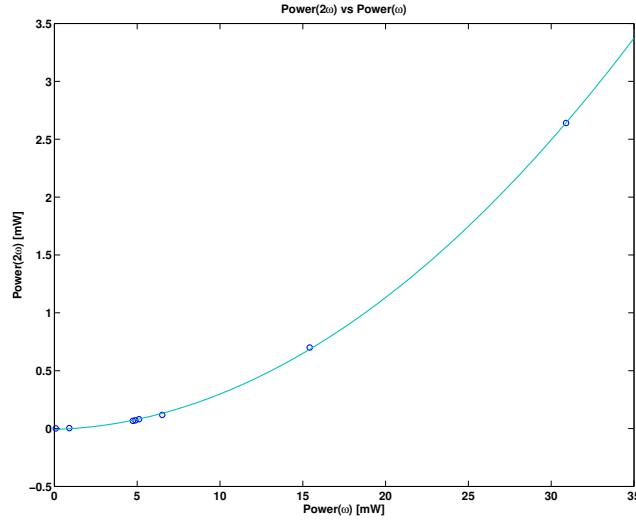
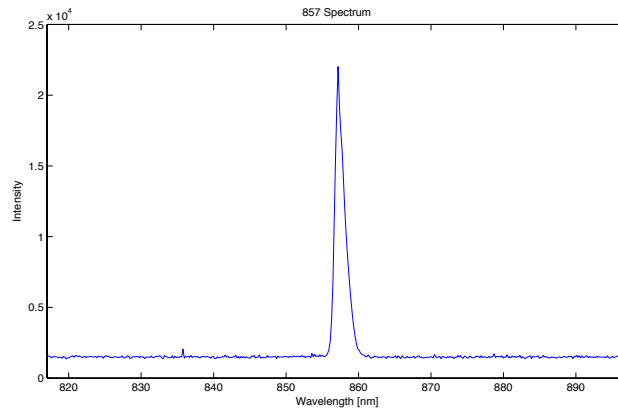


Figure 3.10: measurement of the conversion efficiency of the second harmonic generation setup. The data was taken with a Thorlabs calibrated power meter and CVI neutral density filters. The data is plotted as taken, since both were direct measurements from the calibrated photodiode. The data is well fit by a quadratic as expected.

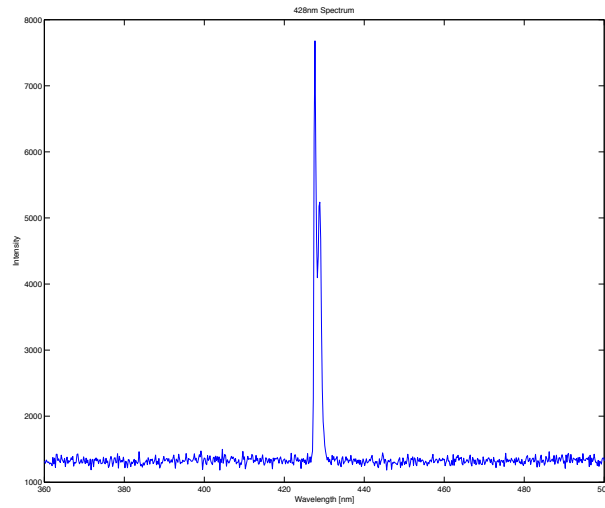
In this case, $f_{eff} = 55.8$ mm for this pair of lenses, using corrected focal distance numbers. The focal distance corrections are discussed below.

A similar process is used to optimize the fourth harmonic generation stage. First, the C2 is placed such that the focus of L4 is within the crystal and L5 is optimized to give a 1mm spot size on the sample. The detector photocurrent is then monitored either directly through return current, or by collection of photoelectrons on the TOF detector. The C2 angle is then tuned to achieve maximum current and therefore satisfy the phase matching condition of that stage. Finally, the L4 to C2 distance can be adjusted to further maximize the photocurrent. The harmonic setup can generate $118\mu W$ of fourth harmonic power and its wavelength verified by an Ocean Optics spectrometer as shown in Figure . Conversion efficiency is again measured by attenuation of the second harmonic beam and monitoring of the fourth harmonic on a photodiode placed after mirror M9. Table 3.3 and Table 3.4 lists the expected conversion efficiency and system parameters for each stage of the conversion process, again resulting in an E2 dependence on energy density.

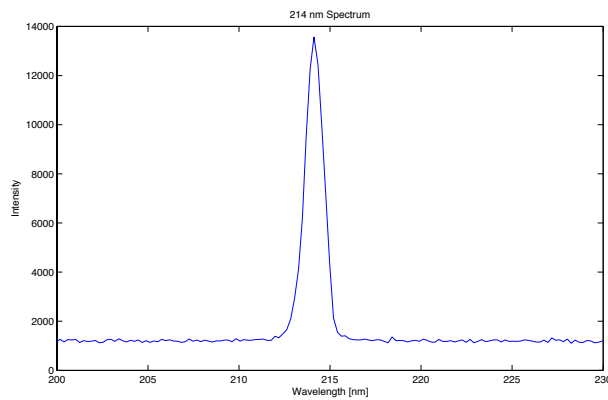
For this setup, stock UV fused silica and BK7 lenses are used. Optical components used far from their design wavelength exhibit modified focal distance due to the change in index of refraction of the lens material at the target wavelength. This phenomena is exhibited as a chromatic focusing, observable after lens L5. As such, it is observed that the focus for the fourth harmonic occurs at a point where the second harmonic is defocused, resulting in two "lines" of second harmonic light (front and back reflections from the polarizing beamsplitter)



(a) Ocean optics spectrum of the fundamental beam from the Oscillator.



(b) spectrum of the output of the second harmonic generation crystal (LBO), taken from a mirror reflection.



(c) spectrum of the 214nm beam taken after rejection of the 432nm beam by two 45 degree polarizing beamsplitters

Figure 3.11: Verification of wavelength operation of the harmonic system.

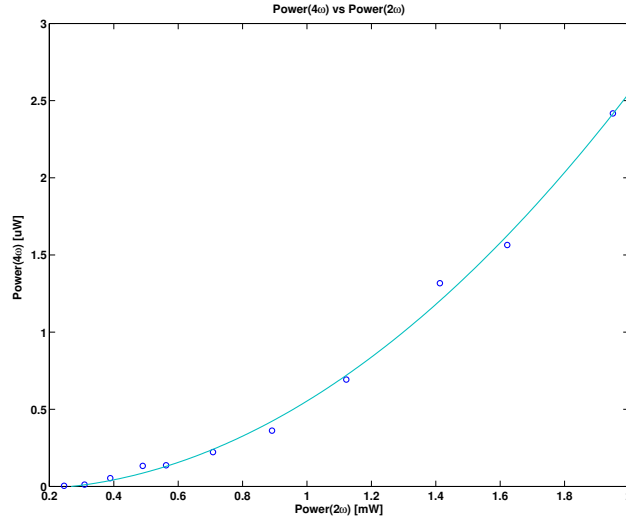


Figure 3.12: The conversion efficiency of the fourth harmonic crystal. The data was taken using the Newport Calibrated UVSi photodiode and the New Focus 5215 Neutral Density Filter wheel to attenuate the 428nm beam. At 2mW, a 0.12% conversion efficiency is observed.

bordering a 1mm spot of focused fourth harmonic light when measured close to mirror M8, less than 500mm from L5. Due to the k-matching condition at crystal C2, the UV spot is emitted at an angle of approximately 2 degrees to that of the second harmonic light, and the UV spot appears to be located closest to the back reflection of the M8 mirror at a distance of 1000mm from L5. Such an effect can be used for coarse alignment of the UV beam on the surface of the sample.

As a metric for compensation for this chromatic focusing, the modified focal distance for each of the lenses used in this system are calculated. Material data on the index of refraction of BK7 and UV Fused Silica were obtained from the Melles Griot Optics guide, and the lensmakers equation under the assumption of a thin lens:

$$\frac{1}{f} = (n - 1) \left[\frac{1}{R_1} - \frac{1}{R_2} + \frac{(n-1)d}{nR_1R_2} \right] \quad (3.3)$$

$$\frac{1}{f} \approx (n - 1) \left[\frac{1}{R_1} - \frac{1}{R_2} \right]$$

for $d \ll R_1, R_2$, was used to calculate the actual focal distance. The chromatic effect was shown to be greater at higher focal distances, or larger deviations from the central wavelength of the lens. This section described the laser and harmonic generation systems of the Photocathode Laboratory. The theory of harmonic generation in nonlinear crystals was discussed and the optics and calibration of the implemented system was presented. Conversion efficiency of the cascaded generation stages was determined to be the most critical

parameter in the implementation. Maximum conversion efficiencies of 20% in the second harmonic stage and 5% in the fourth harmonic stage were measured. The resulting system is tunable in the UV. By adjusting the fundamental beam wavelength and the angle of the crystal provides sufficient photon flux for the DLD TOF detector.

Table 3.5: System parameters and expected performance of the Second Harmonic stage (857 nm - 432 nm)

Parameter	Value
857 nm Beam size/ shape	1 mm round (M3)
L1 to C1 distance	5.5 cm
C1 to L2 distance	4.5 cm
C1 crystal angle (θ, ϕ)	(265,3)
2ω Maximum Conversion efficiency (measured)	20%, 2.4mW

Table 3.6: System parameters and expected performance of the Fourth Harmonic stage (432nm - 214nm)

Parameter	Value
423 nm Beam size/ shape	2 mm round (M5)
L3 to M5 distance	8.8 cm
L4 to C2 distance	6 cm
C2 crystal angle (θ, ϕ)	(63,121)
4ω Maximum Conversion efficiency (measured)	5%, 118 μ W

3.5 The Fowler Method of Work Function Measurement

The work function of a surface is a critical parameter in determining the electron emission characteristics and is an exquisitely sensitive tool in assessing sample cleanliness; often a single monolayer of a gas adsorbed on a surface can cause work function changes of up to 1 eV.[12] This section describes the construction of an optical system to measure the sample work function by the Fowler method. Details of the extraction of the workfunction are presented in the next chapter.

Figure 3.13 and Figure 3.14 show a schematic and a photograph of the Fowler system. An Energtiq EQ-1500 laser plasma source is used as a broadband CW source (170 nm - 800

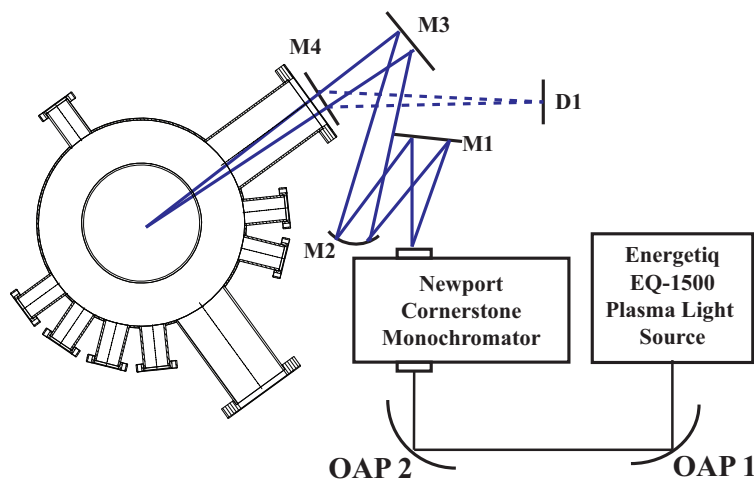


Figure 3.13: A schematic layout of the Fowler system components. The Energetiq laser plasma light source is focused into a 1:1 Ebert-Fastie monochromator (Newport). The monochromatized beam is then focused onto the sample by a folded arrangement (M1 - M3). A flip mirror, M4, can be optionally introduced to reflect the focused beam on photodiode D1, to measure the photon flux delivered to the sample. This folded cavity system fits on a 600 mm x 300 mm optical bench.

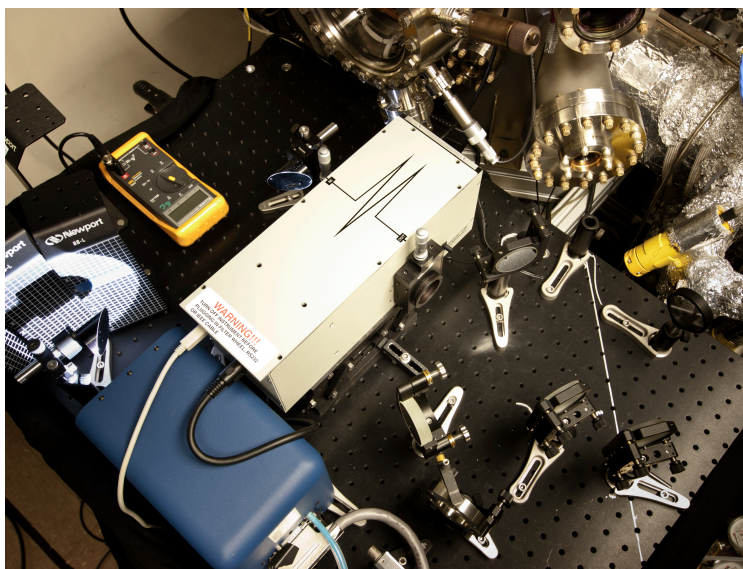


Figure 3.14: A photograph of the optical bench comprising the light source for the Fowler measurement. The laser plasma source is focused into a Newport monochromator using two 5mm focal length Off Axis Paraboloids. The monochromatized beam is then collected and focused onto the sample using MgF2 overcoated, Al-coated curved mirrors.

nm) and is focused into a LabView controlled Newport monochromator by two replicated off-axis paraboloid mirrors. After the monochromator, the light is then focused in a 1mm spot on the sample by a folded optical cavity, comprised of two flat and one folded mirror. A third flip mirror can be optionally introduced into the optical path and directs the beam onto a calibrated photodiode to measure the flux delivered to the sample. Figure 3.16 and Figure 3.17 show the photon flux at the exit of the monochromator and the monochromator calibration, respectively.

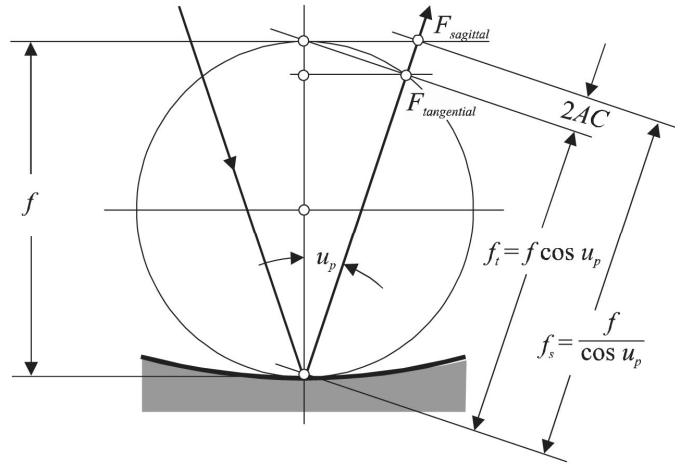


Figure 3.15: A depiction of the relationship for the longitudinal distance between the tangential and sagittal foci of a spherical mirror for a beam with a central ray at half-angle u_p . [13]

To avoid chromatic defocusing of the sample spot, a single spherical curved mirror was used to image the exit slit of the monochromator to on the sample. The condition for 1:1 imaging by a curved mirror is given by [14]:

$$\frac{1}{f} = \frac{1}{S_1} + \frac{1}{S_2} \quad (3.4)$$

where f is the focal distance of the mirror and S_1 , S_2 are the object and image distances, respectively. For a beam incident at a non-normal angle, the transverse foci are separate longitudinally. This separation together with the convergence angle gives an astigmatic blurring at some intermediate plane; reduction of this blur to a tolerable level depends on the angle of incidence, magnification and convergence angle. In order to calculate this, we start with the equations describing the tangential and sagittal focal conditions, where S_1 is the object distance, S_2 and S_3 are the tangential and sagittal image distances, R is the mirror radius, and U_p is the angle of incidence. The relationship of the focal lengths is shown

Table 3.7: A Catalog of Optics and Instrumentation used in the Fowler System

	Manufacturer/ Part Number	Specification	f [mm]
OAP1	Newport 50338AL	Replicated Off Axis Paraboloid	101.6
OAP2	Newport 50338AL	Replicated Off Axis Paraboloid	101.6
M1	CVI DUVA-SMCC-2037-0.50-UV	50.8mm diameter, 1/10 Wave, AL mirror	-
M2	CVI DUVA-PM-2037-UV	Concave Mirror, 50.8mm diam, AL Coating	250 mm
M3	CVI DUVA-SMCC-2037-0.50-UV	50.8mm diameter, 1/10 Wave, AL mirror	-
M4 (Flip)	CVI DUVA-SMCC-2037-0.50-UV	50.8mm diameter, 1/10 Wave, AL mirror	-
D1	IRD AXUV100 O3-2	Calibrated UV Photodiode	-
-	Newport Cornerstone USB Monochromator	70023 Ruled Grating (Blaze 250nm) 70022 Holographic Grating (Blaze 350nm)	-
-	Energetiq EQ-1500 Laser Plasma Source		

in Figure 3.15. Accounting for this relationship in 3.4 yields two equations:

$$\frac{1}{S_1} + \frac{1}{S_2} = \frac{2}{R_t \cos(u_p)} \quad (3.5)$$

$$\frac{1}{S_1} + \frac{1}{S_3} = \frac{2 \cos(u_p)}{R_s} \quad (3.6)$$

where R_s and R_t are the radii of curvature of the mirror in the tangential and sagittal directions, related to the mirror focal length by:

$$f = \frac{R}{2} \quad (3.7)$$

For a spherical mirror, the radii of curvature are equivalent, and $R_s = R_t$. Additionally, for one-to-one imaging, $S_1 = S_2$. The two equations can be combined into a single

relationship relating the image distances and the angle of incidence:

$$2 \cos(u_p) - 1 = \frac{S_1}{S_3} \quad (3.8)$$

For a 1mm image, an additional 1mm for astigmatism, and an angular convergence of $6 = 100$ mrad:

$$|S_1 - S_3| \leq \frac{1mm}{100mrad} \quad (3.9)$$

For a image distance of $[S_1] = 500\text{mm}$, (6.8) requires $[S_3] = 510$ mm and (6.7) predicts an incidence half angle of $[U_p] = 5.7$ degrees. At this angle of incidence and a beam diameter which fills the mirror ($b = 50\text{mm}$), the minimum distance between optics, l , can be approximated by:

$$l \simeq \frac{b}{\sin(2u_p)} \quad (3.10)$$

For the parameters as defined here, $l = 253$ mm. The measured photocurrent as a function of wavelength is normalized to the incident photon flux through the optical system. This correction factor must also include the wavelength dependent reflectivity of the components in the optical path. Analysis of the resulting sample photocurrent as a function of wavelength data is discussed in Chapter 5.

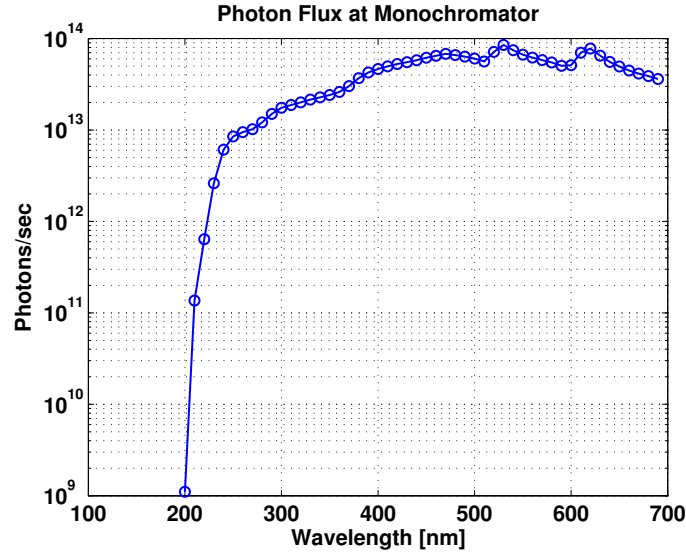


Figure 3.16: The measured flux [photons/sec] as a function of wavelength at the exit of the monochromator as measured by the calibrated AXUV photodiode.

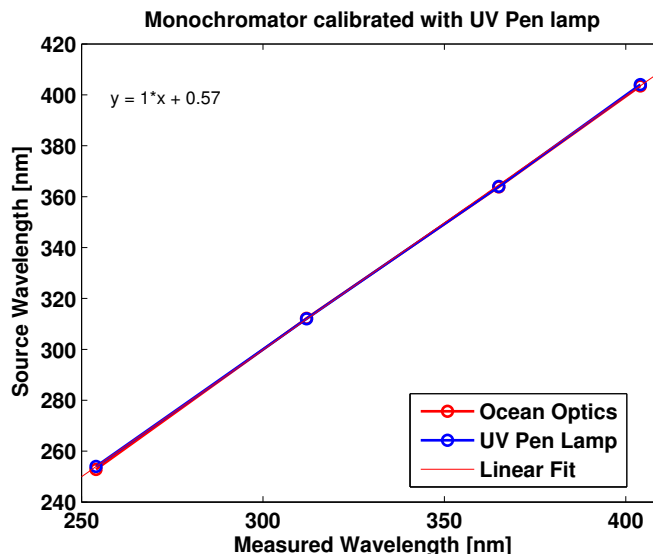


Figure 3.17: Calibration of the monochromator using the peak emissions of a tungsten UV Pen lamp.

3.6 Time of Flight Angle Resolved Photoemission Spectroscopy (TOF-ARPES)

Table 3.8: Surface Concepts DLD 4242 detector parameters [17]

Detector Parameter	Specification Value
Diameter:	44 mm
Number of Pixels:	X: 1389 Y: 1484
Pixel Size:	X: 31.7m Y: 29.7m
Spatial Resolution (Rayleigh):	3-4 pixels
Dark Counts:	5 counts/sec
Time Resolution:	144 ps
MCP Bias Voltage:	2.5 kV

Chapter 2 outlined the three fundamental procedures of the Three Step Model of Photoemission to be investigated in the Photoemission Laboratory. The model predicts an continuous distribution of electron momenta outside of the photocathode. In order to investigate this prediction, the electron momenta are measured using Angle Resolved Photoemission Spectroscopy (ARPES). In this section, the ARPES concept is developed, and the detector

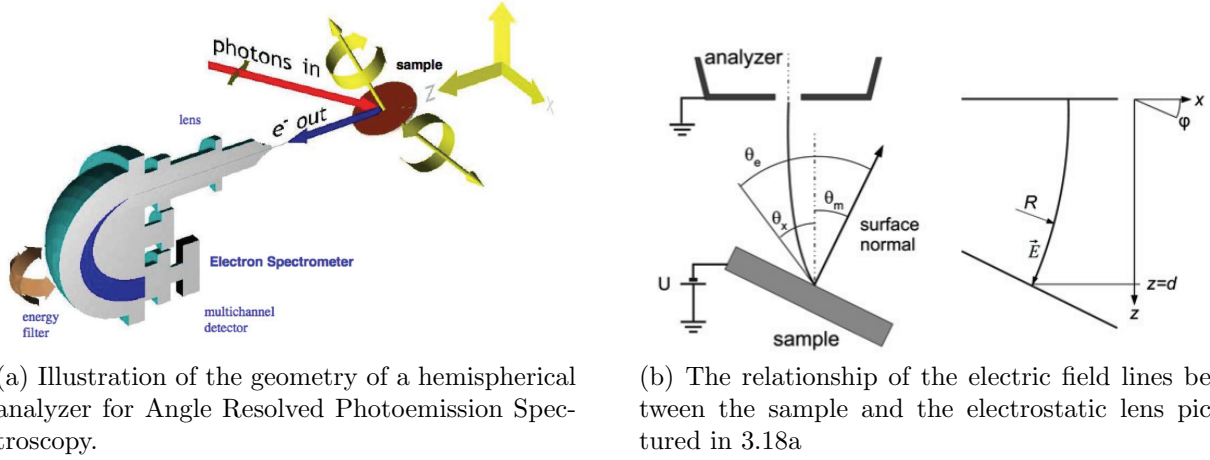


Figure 3.18: A depiction of sample geometry and electric field considerations in Angle Resolved Photoemission Spectroscopy. The electrostatic lens images electrons from the sample to the entrance slit of the analyzer. The detector records the number of electron counts as a function of energy and angle. To investigate the entire Brillouin zone, the sample is rotated.[15, 16]

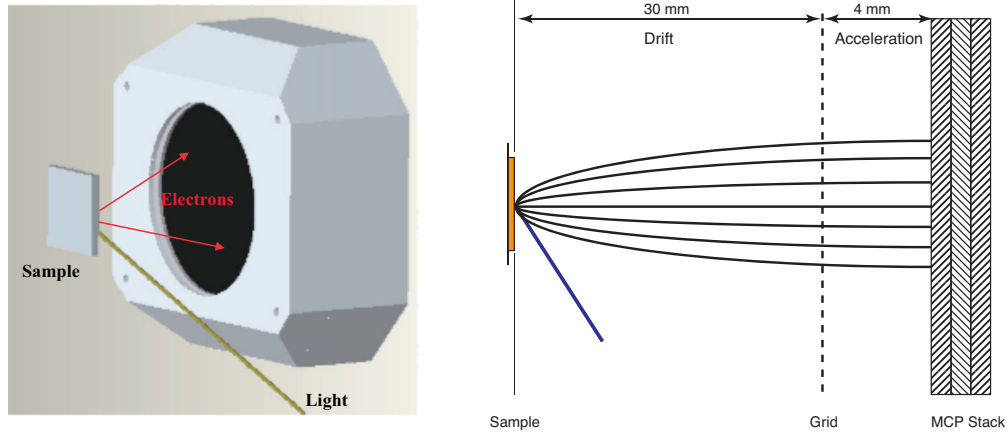


Figure 3.19: Represents the parallel plate sample-detector geometry. The sample plane sits 30mm in front of the accelerating grid. The laser enters at a 39 degree angle of incidence to the plane of the sample. The photoemitted electrons follow parabolic trajectories to the MCP stack. Accelerating bias voltages are applied to the sample, grid and MCP stack independently.

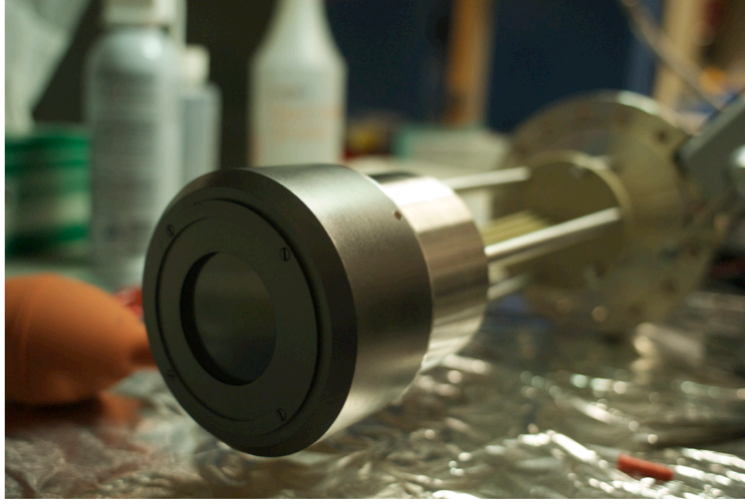


Figure 3.20: A photograph of the in-vacuum detector head. Electrons are incident on the MCP through the grid and the out-of-vacuum electronics chain processes resulting pulses incident on the meander lines.

and relevant details of the experimental setup are presented.

Figure 3.20 shows the typical electron analyzer setup for collection of angle resolved datasets. A sample is illuminated by an external photon source. Photoemitted electrons are collected and focused onto the entrance slit of a biased hemispherical capacitor by an electrostatic lens.[15] Only those electrons that satisfy a specific pass energy E_{pass} are accepted into the hemispherical region. The hemispherical path disperses electrons spatially according to the vectorial components of their energy; Electrons of higher energy follow paths of larger radii through the hemisphere than those of lower energy. The resulting distribution on a detector at the exit of the hemisphere records the number of electrons as a function of sample angle and transverse energy, for a total energy window centered at E_{pass} . The full energy spectrum of the electrons can then be evaluated in multiple exposures by scanning E_{pass} , the hemisphere bias voltage and the complete Brillouin zone is investigated by rotation of the sample.

The near-band edge photoemission under investigation produces near zero kinetic energy electrons at the surface of the cathode with the maximum energy being < 1 eV. These electrons are particularly susceptible to acceleration by stray electric and magnetic fields within the experimental chamber from the analyzer. The ideal analyzer is a 2-dimensional spatially resolved (x,y) parallel plate geometry, in which the accelerating field is static and perpendicular to the surface of the sample. The parallel component of the electron momentum is unaffected by the accelerating field; the tangential momentum is increased by the imposed bias across the two plates, multiplied by the electronic charge. Energy resolution is achieved by measuring the time of arrival of the electron over a fixed distance d , at the

analyzer, relative to a starting time zero (t_0), such that [18]:

$$E_{kin} = \frac{m_e}{2} \left(\frac{d - d_0}{t - t_0} \right)^2 \quad (3.11)$$

The angle of photoemission θ , is given by the (x,y) coordinate of the electron at the analyzer through:

$$\hbar k_{\parallel} = \sin(\theta) \sqrt{2m_e E_{kin}} \quad (3.12)$$

where

$$k_{\parallel} = \sqrt{k_x^2 + k_y^2} \quad (3.13)$$

and

$$k_{(x,y)} = \frac{m_e}{\hbar} \left(\frac{x - x_0}{t - t_0} \right) \quad (3.14)$$

This is termed the Time of Flight (TOF) method of electron spectroscopy.[19] Figure 3.19 displays the experimental geometry. The TOF analyzer chosen, shown in Figure 3.20, is a two-dimensional Delay Line Detector constructed by Surface Concepts, GmbH. Consisting of an in vacuum detector head and external detector electronics, the detector measures position (x,y) and the time of arrival of an electron pulse, relative to a laser-derived trigger.[17] The specifications of the detector are given in Table 8.

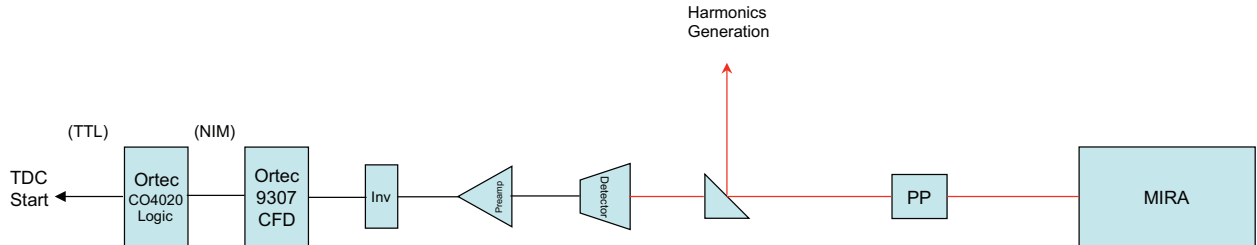


Figure 3.21: A schematic depiction of the generation of the DLD trigger from the pulse-picked signal. A biased silicon photodiode captures light leakage from a steering mirror and produces a signal then sent to a preamplifier. Red lines indicate optical signals; black line are electrical signals. The diode output is inverted, compared to a triggering threshold in a constant fraction discriminator (Ortec 9307) and then an appropriate TTL trigger pulse is generated. Figure 3.22 shows the relative timing of the signals presented here.

The detector trigger synchronization signal is derived from a photodiode monitoring the leakage of the fundamental beam from steering mirrors at the exit of the pulse picker, as shown in Figure 3.2. This 4MHz signal is converted to a TTL logic pulse using a constant fraction timing discriminator and a NIM to TTL Logic unit. The electronics chain and timing diagram for each component are shown in Figure 3.21 and Figure 3.22, respectively.

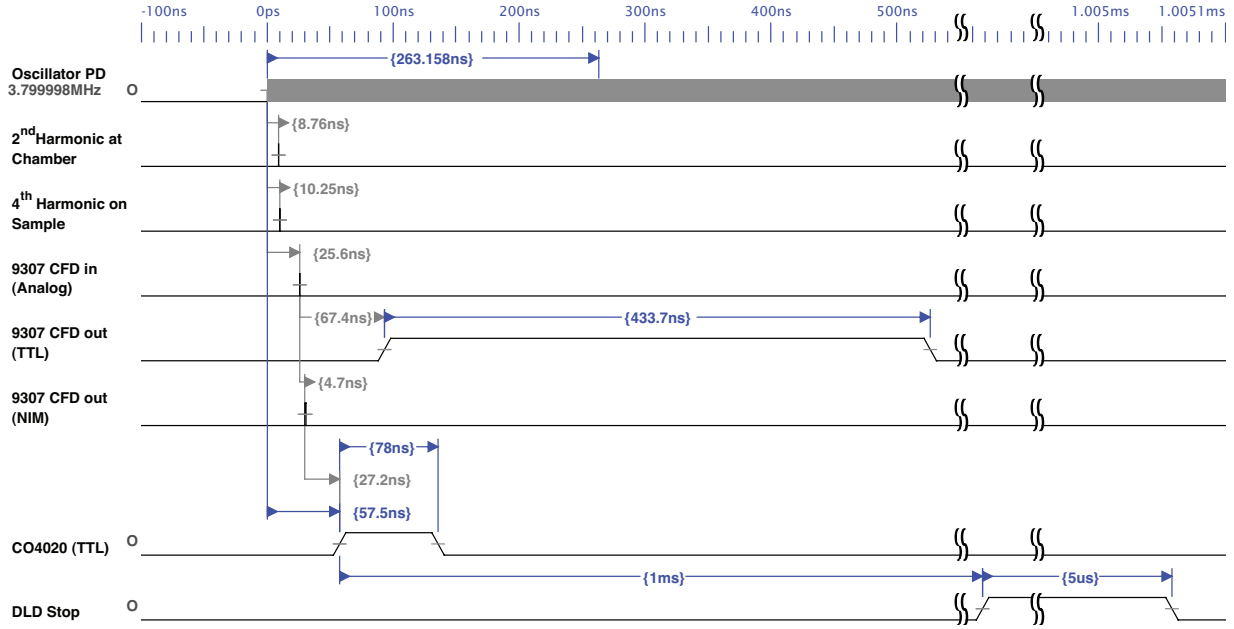


Figure 3.22: A timing diagram detailing the triggering of the TOF detector relative to the oscillator output.

The trigger synchronizes the detector to the signal on the microsecond timescale. Within the detector software, a programmable fine delay can be set to adjust the time window to nanosecond precision. The time window duration is limited to 40ns; integration times of milliseconds are integrated by repeated samples of the time window. The DLD analyzer is controlled via USB using a Labview API from the experiment PC. Calls to the vendor-produced API are interfaced using a customized C library and Labview code interface nodes. Prototyping of the C-based analysis was done in MATLAB. Both the MATLAB codes and the C-library are included in the appendices.

3.7 Sample Preparation and Characterization

Investigation of the electronic properties of metallic photocathodes requires the preparation of the single crystalline material under UHV conditions. Czochralski grown samples of Cu(111) and (100) oriented crystals were purchased from Surface Preparation Laboratory. The sample dimensions are 10mm in diameter and 1mm thick. A thermocouple hole of diameter 0.5mm is spark eroded along the height, as are two slits for mounting the crystal to the sample holder. The face of the sample is polished to a surface roughness of 0.03 micron.

The samples are then introduced into the vacuum system and undergo repeated cycles

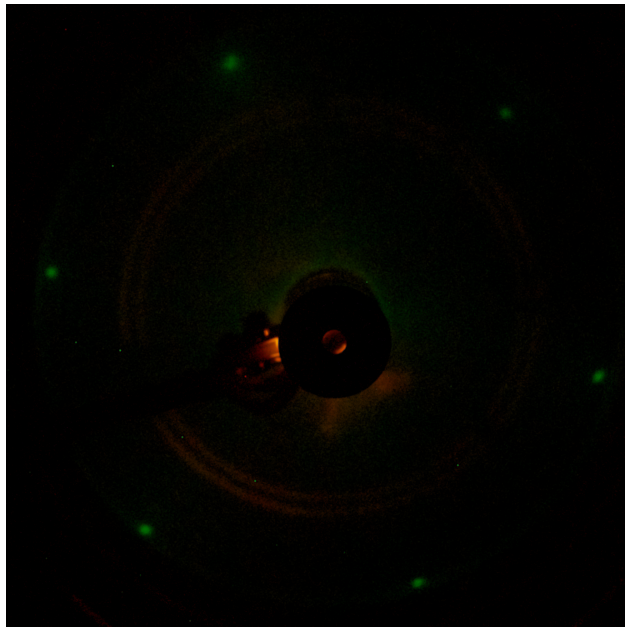


Figure 3.23: A representative LEED spectrum of a clean (111) oriented Copper sample after repeated ion sputtering and annealing cycles. The pattern is taken at electron energy of 79.3 eV.

of ion sputtering and sample annealing. A SPECS IQE-11 Ion gun system is operated at a chamber pressure of 5×10^{-5} Torr of Argon at an ion energy of 1kV. The accelerated Argon ions impact the surface of the sample at an angle of 45 to the surface normal and remove 10 of copper during the 15 minute sputtering cycle. The samples are then annealed at 500° C for 2 hours to reconstruct the crystalline lattice at the surface. After cooling to 30°C, low energy electron diffraction (LEED) was used to verify the crystalline orientation. Sputtering and annealing cycles were repeated until both a strong LEED pattern was achieved and the sample work function as measured by the Fowler method matched published values.[20, 21] A representative LEED pattern is shown in Figure 3.23.

3.8 Sample Biasing and Photocurrent Measurement

A fundamental requirement for all experiments in the photoemission chamber is the biasing of the sample relative to the chamber walls and the analyzer potential. A Keithley 6517B Electrometer / Voltage Source was used to both provide the bias voltage and measure the sample photocurrents. This instrument is capable of sourcing 1kV potential at 10mA and measuring dc currents from 10 aA to 21mA. The electrometer was configured in "Force Voltage Measure Current" mode, in which the ammeter and the voltage source are connected in series with the sample, R_L . [22] This configuration is shown in Figure 3.24. The electrome-

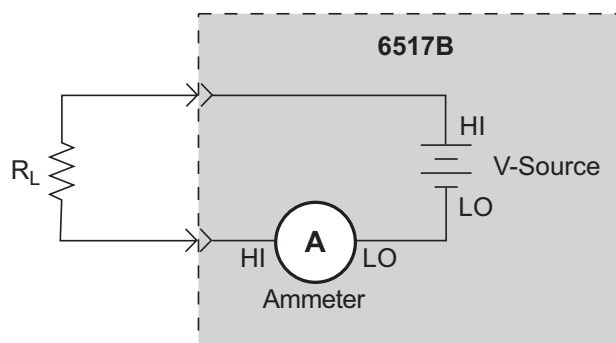


Figure 3.24: The circuit connection of the picoammeter and internal voltage source (Keithley 6517B) used to make photocurrent measurements.[\[22\]](#)

ter is configured and controlled by GPIB commands using Labview codes on the experiment PC.

Chapter 4

Data Collection, Analysis and Results

4.1 Introduction

The Photoemission Characterization Laboratory was built in order to characterize the quantum yield of material systems and the transverse emittance of the resulting photoemitted electron beams. As discussed in previous sections, metal photocathodes are of particular interest due to their fast response, ease of fabrication, long lifetime and operation in the $\times 10^{-9}$ Torr regime due to their vacuum stability.

As a starting point for these investigations, the first polycrystalline copper photocathode commissioned in the LCLS RF gun was removed and analyzed on Beamline 12.3.2 of the Advanced Light Source (ALS) using white-light X-ray scanning microdiffraction (SXRD). The XRD data shows that the LCLS photocathode consists of a single-crystal grain orientation within the drive laser beam size. The result of this investigation motivated the use of single-crystal copper photocathode substrates for further analysis.

Studies of single crystal copper samples of (100) and (111) orientations were completed in the Photoemission Characterization Laboratory. Prior to evaluation, the surface of each crystal was prepared by repeated ion-bombard and anneal cycles. The cleaning cycles were repeated until the material work function matched values previously reported in the literature as measured by the Energy Dependent Yield (EDY) and fit using the Fowler method. As a second check of material order, a Low Energy Electron Diffraction (LEED) measurement was conducted in order to verify the crystalline orientation.

Once prepared, the samples were then evaluated to investigate the relevant assertions of the three-step model of photoemission. The dependence of the sample quantum yield on the angle of incidence of the incoming light was studied as a function of photon energy (AREY). Finally the momentum distribution of the photoemitted electrons was measured using Time of Flight Angle Resolved Photoemission Spectroscopy (TOF-ARPES). The measurement details, data analysis and results of these investigations are presented in this chapter.

To assess the degree to which ion backscattering and surface order impact the quantum yield and emittance of electron beams created using single crystal photocathodes, exper-

iments were conducted to correlate surface order and quantum yield. The result of this investigation is reported. Finally, the influence of the sample electronic structure on the quantum yield and the emittance of the resulting electron beam are discussed at the end of the chapter.

4.2 Current Photocathodes: Characterization of Crystallinity by Scanning X-Ray Microdiffraction

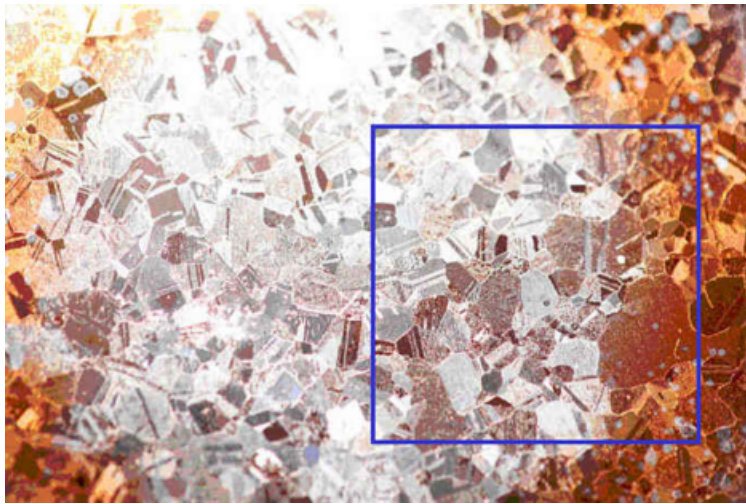


Figure 4.1: A white light image of the center of the LCLS photocathode. The horizontal dimension in this image is 30mm. The structures of the surface are shown to be grains of single crystal orientation. The typical grain size is 1 - 2 mm; comparable to the size of the laser spot in LCLS. The image was taken with a Canon EOS T1i digital camera with a 60 mm lens and using flash illumination. The 1 cm x 1 cm blue square represents the region of interest for the study described here.

Figure 4.1 shows a white light photograph of the first commissioned photocathode from the LCLS Free Electron Laser at Stanford. The surface of the photocathode is polycrystalline in nature.[4] White light scanning X-ray microdiffraction on Beamline 12.3.2 of the Advanced Light Source was used to identify the orientations of the polycrystalline grains. A bright field image at 5x magnification of the region of interest in Figure 4.1 is shown in Figure 4.2. In this figure, the contrast has been adjusted to highlight the grain and twin boundaries evident on the surface of the photocathode.

The region of interest was then studied by scanning X-Ray microdiffraction with a 5-22 keV white beam from the ALS. The 1 cm x 1cm area was scanned with a step size of $100\mu\text{m}$ x $100\mu\text{m}$ and a beam size of $1\mu\text{m}$. At each point a Laue pattern was collected and

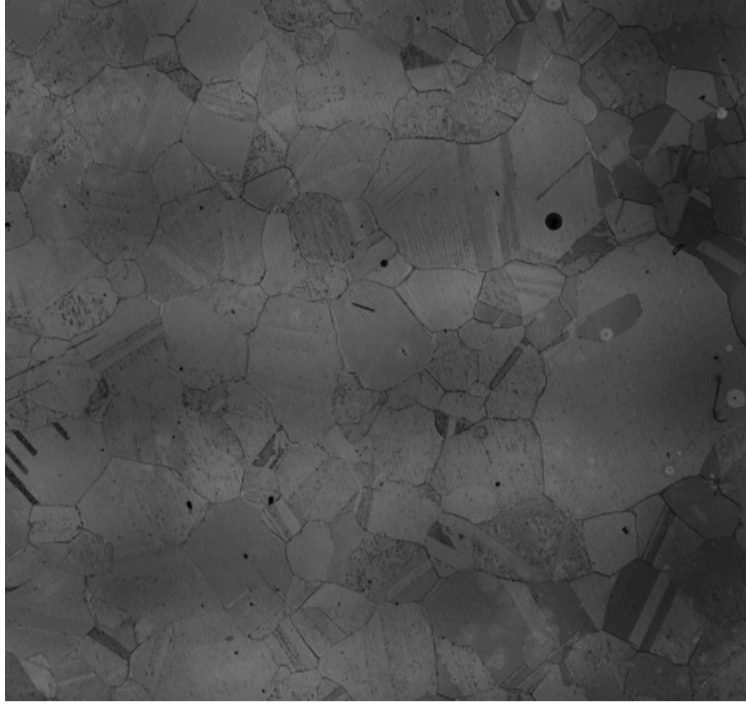


Figure 4.2: The bright field image of the region of interest pictured in Figure 4.1 taken at 5x magnification. The imaged area is 1 cm x 1 cm. The image contrast has been adjusted to highlight the grain and twinning boundaries evident on the surface of the photocathode.

indexed to identify the crystalline orientation at that point. A representative Laue pattern from a single point in the scan is shown in Figure 4.3. In this figure, the indexed reflections appear as streaks, differing in intensity. Points of high intensity (red) are reflections from the surface of the sample; the lower intensity points from beneath the sample surface. Such behavior indicates that the crystalline orientation of the surface and the bulk differ. In order to confirm this, 3D X-Ray microdiffraction measurements are required.

Figure 4.4 shows the region of interest of Figure 4.2, indexed by grain orientation as represented by the color index shown. The sample surface consists of (111) and (100) oriented grains as indicated by blue and red shading, respectively. The range of sizes of the polycrystalline grains are approximately 1-2 mm. Dependent on the size of the illuminated spot, photoemission from such a cathode is dominated by a single crystal grain with a defined work function. For the LCLS photogun, the spot size is 1mm, indicating that this is the case. Furthermore, because the quantum yield is a function of the excess energy of the impinging photon, illumination of different crystalline orientations on the photocathode would produce different quantum yields. The work functions of Cu(111) and Cu(100) are 4.94 eV and 4.59 eV, respectively.[19] Given the drive laser wavelength and accelerating gradient (20 MV/m) present in the LCLS photogun, the excess energies of the (111) and (100) orienta-

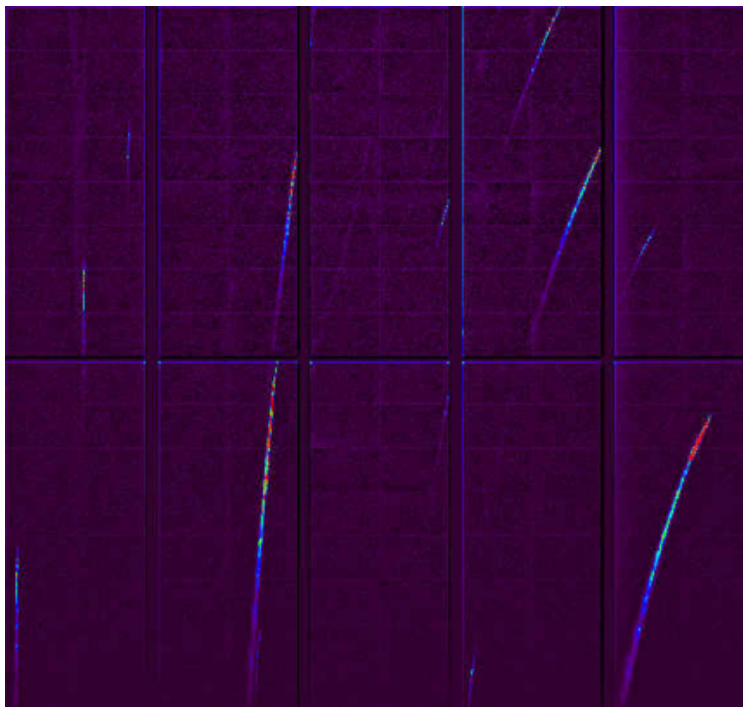


Figure 4.3: A representative Laue pattern from a single point of the SXRD experiment. In this diffraction pattern, the angular range of one Laue peak is 8 degrees. The top and most intense part of the reflections come from near the surface; the bottom is from deeper within the sample. The streaking of the diffraction spots indicates misorientation of the top surface and bulk grains, most likely introduced in the polishing process.

tions are 0.1 eV and 0.45 eV, respectively. The Three-Step Model of Photoemission predicts a difference in yield of a factor of 20, dependent on the work function of the illuminated grain. Furthermore, the emittance of the electron beam exhibited between the Cu(111) and Cu(100) oriented grains is very different, due to the relationship of quantum efficiency to photon excess energy. Therefore, an understanding of the overall performance of a metal photocathode is helpful in achieving a low emittance, high QE, photocathode. This result of this study motivates the investigation of the predictions of the Three Step model in the context of single crystal metal photocathodes. These investigations were conducted within the photoemission laboratory and are reported in the sections to follow.

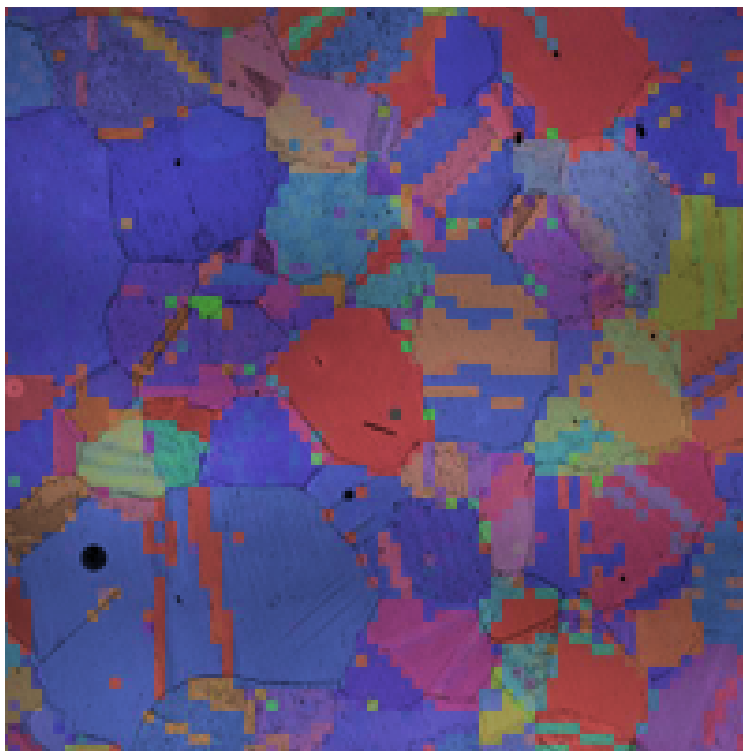


Figure 4.4: An overlay of Figure 4.2 and the RGB uSXR orientation map of the LCLS Photocathode. The field of view is 20 mm x 26 mm. Blue points indicate regions of (111) orientation; Red points indicate regions of (100) orientation, and Green points are of (100) orientation. The scanned area consists predominately of (111) and (100) oriented grains; indicating that the overall work function of the sample is dominated by the work function of these two crystalline orientations.

4.3 Single Crystal Studies: Determination of the Work Function by the Fowler Method

In order to verify that the prepared sample was of a single crystalline orientation, two methods were employed. First, the Low Energy Electron Diffraction pattern (LEED) was recorded. As a second check, the work function of the surface was evaluated by measuring the sample current as a function of photon energy and the data fit by the Fowler method.[4, 23]

Figure 4.5 depicts the photoemitted current measured by the sample as a function of the incident photon energy. From this plot, the work function threshold is not readily apparent. The Fowler method describes a functional form of the Photoelectric current versus photon energy, from which the work function can be determined. The Fowler method makes three fundamental assumptions, namely:

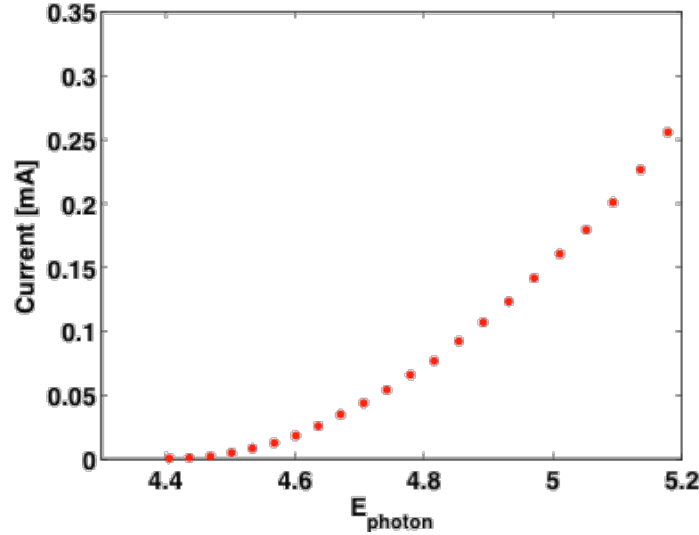


Figure 4.5: The photoelectric current emitted from the sample as a function of photon energy. The data is fit using the Fowler method in order to extract the sample work function.

1. Only those electrons with sufficient kinetic energy perpendicular to the surface contribute to the photoelectric current.
2. The number of electrons available for photoemission, N_B , scales proportionally with T^2 , for temperature T .
3. The density of states of the material near the Fermi level is constant.

The derivation of the fitting function proceeds with the assumption of Fermi-Dirac distributed electron availability as detailed in Fowler 1963. [23] According to Fermi statistics, the number of available electrons per unit volume,

$$n(u, v, \omega) \quad (4.1)$$

is given by:

$$n(u, v, \omega) du dv d\omega = 2 \left(\frac{m}{h} \right)^3 \frac{du dv d\omega}{e^{\left(\frac{m}{2} (u^2 + v^2 + \omega^2) - E_F \right) / k_B T} + 1} \quad (4.2)$$

Integrating out the spatial dimensions, Equation is reduced to express the number of available electrons in energy range u to $u+du$:

$$\tilde{n}(u) du = \frac{4\pi k_B T}{m} \left(\frac{m}{h} \right)^3 \log \left\{ 1 + e^{\left(E_F - \frac{mu^2}{2} \right) / k_B T} \right\} du \quad (4.3)$$

where E_F is the material Fermi level. The thermionic work function ϕ is related to the Fermi level and the total potential barrier height χ_0 by: $\phi = \chi_0 - E_F$. The total number of photoemitted electrons, N_B , is given by:

$$N_B = \int_{\chi_0 - h\nu}^{\infty} \tilde{n}(u) du \approx \left(\frac{2\sqrt{2}\pi m^{3/2}}{h^3} \right) \left(\frac{k_B^2 T^2}{(\chi_0 - h\nu)^{1/2}} \right) \int_0^{\infty} \log \{1 + e^{-y + (h\nu - \chi)/k_B T}\} dy \quad (4.4)$$

In order to complete the integration in Equation , the logarithm is expanded term-by-term and integrated. This results in two distinct intervals of integration for $\mu = \frac{h\nu - \chi}{k_B T}$: For $\mu \leq 0$:

$$N_B = \left(\frac{2\sqrt{2}\pi m^{3/2}}{h^3} \right) \left(\frac{k_B^2 T^2}{(\chi_0 - h\nu)^{1/2}} \right) \left[e^{\mu} - \frac{e^{2\mu}}{2^2} + \frac{e^{3\mu}}{3^2} - \dots \right] \quad (4.5)$$

and for $\mu \geq 0$:

$$N_B = \left(\frac{2\sqrt{2}\pi m^{3/2}}{h^3} \right) \left(\frac{k_B^2 T^2}{(\chi_0 - h\nu)^{1/2}} \right) \left[\frac{\pi}{6} + \frac{\mu^2}{2} - \left\{ e^{-\mu} - \frac{e^{-2\mu}}{2^2} + \frac{e^{-3\mu}}{3^2} - \dots \right\} \right] \quad (4.6)$$

Under the third assumption listed above, the photoelectric current I is proportional to the number of electrons contributing to the photoelectric current, such that $I \propto N_B \propto T^2$. The terms in front of the expansions in Equations and can then be collected in a single coefficient A , such that:

$$\frac{I}{T^2} = Af \left(\frac{h\nu - \phi}{k_B T} \right) \quad (4.7)$$

where:

$$f(\mu) = \begin{cases} e^{\mu} - \frac{e^{2\mu}}{2^2} + \frac{e^{3\mu}}{3^2} - \dots & (h\nu \geq \phi) \\ \frac{\pi}{6} + \frac{\mu^2}{2} - \left\{ e^{-\mu} - \frac{e^{-2\mu}}{2^2} + \frac{e^{-3\mu}}{3^2} - \dots \right\} & (h\nu < \phi) \end{cases} \quad (4.8)$$

Expressing Equation in terms of logarithms reveals fit constants B and ϕ in the Fowler relation:

$$\log \left(\frac{I}{T^2} \right) = B + \log \left(f \left(\frac{E_{\text{photon}} - \phi}{k_B T} \right) \right) \quad (4.9)$$

The resulting fit to the functional form in Equation 4.9 for the data of Figure 4.5 is shown in Figure 4.6, where Equation 4.9 is expanded to three terms in the series. This functional form provides excellent agreement to the data, permitting the extraction of the sample work function from measurement of the photoelectric current vs. wavelength plot.

4.4 Single Crystal Studies: Angle-Resolved Electron Yield (AREY)

In order to quantify the dependence of the sample quantum yield as a function of the angle of incidence of the incoming beam, the Angle-Resolved Electron Yield (AREY) of the

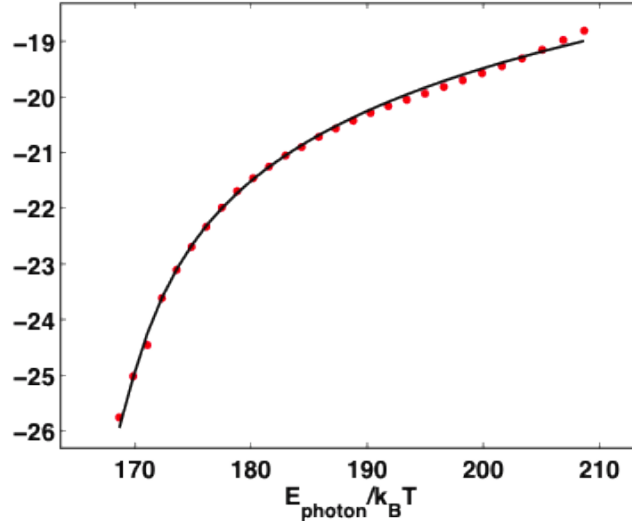


Figure 4.6: The fit of the functional form derived in Equation 4.9 on the data shown in Figure 4.5.

sample was measured. The experimental geometry has been previously shown in Chapter 3. In order to account for the change of the predicted photocurrent as a function of angle of incidence, modifications to step one (absorption of the photon) and step two (the electron escape probability) must be employed. In this section, the relevant equations are derived to account for analysis of the angle of incidence of the incoming beam in the three-step model. Figure 4.7 depicts the coordinate system at the metal-vacuum interface referred to in this section.

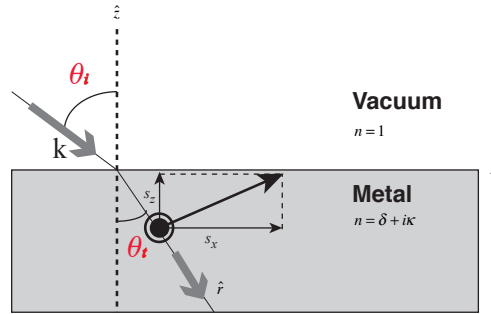


Figure 4.7: The coordinate system at the metal-vacuum interface. n is the index of refraction in each medium. The incident wave is evaluated in the x - z plane ($y=0$).

Modification to Step 1: The Transmission Coefficient

Step 1 of the Three-Step model describes the absorption of the incoming beam through the use of the Fresnel Equations at the metal-vacuum interface. The form of the transmission coefficient used in this step must account for the change in flux per unit area of the incident beam as a function of the angle in the presence of a single complex index of refraction, describing both the bulk and surface material. [24, 6] The form of the transmission coefficient T , derived under these conditions is given by [6]:

$$T = |\sqrt{\epsilon}|^2 \operatorname{Re} \left(\frac{\sqrt{\epsilon - \sin^2(\theta_i)}}{\epsilon \cos(\theta_i)} \right) \left| \frac{2 \cos(\theta_i) \sqrt{\epsilon}}{\epsilon \cos(\theta_i) + \sqrt{\epsilon - \sin^2(\theta_i)}} \right|^2 \quad (4.10)$$

where $n = \sqrt{\epsilon\mu}$ and $\mu = 1$ within the optical range of wavelengths.

Modification to Step 2: The Electron Escape Probability

Step 2 calculates the probability of electron escape from the material using the optical absorption length and the electron mean free path. Both quantities require modification to account for the changing angle of incidence of the incoming beam. First, the optical absorption length as a function of angle of incidence is derived in the case of a lossy metal material with complex index of refraction. Next, the angle of refraction as function of the angle of incidence are derived, from which a form of Snells law is evident. Finally, the angular response of each of the three steps of the model is evaluated for both Copper and Silver (111) photocathodes.

Step 2 of the model predicts the probability of electron escape under the influence of electron-electron scattering (F_{e-e}):

$$F_{e-e} = \frac{1}{1 + \frac{\lambda_{opt}}{\lambda_{e-e}}} \quad (4.11)$$

where λ_{opt} is the optical absorption length within the metal, and λ_{e-e} is the electron-electron mean free path. Matching of the electron mean free path and the optical absorption length as described in Equation 4.11 results in a maximum emission probability of escape for electrons leaving the metal-vacuum interface. The electron mean free path, λ_{e-e} is given by measurements at near-threshold energies. It scales as $E^{3/2}$ and is 2.2 nm at 8.6 eV from the bottom of the valence band.[4] The optical absorption length is given by the imaginary part of the index of refraction of the metal at normal incidence. This length is shortened geometrically by the change in the angle of incidence of the incoming wave. In order to calculate $\lambda_{opt}(\theta_t)$, we use Snells law to calculate both the angle of refraction within the metal and the angular index of refraction.

$$\sin(\theta_t) = \frac{\sin(\theta_i)}{\hat{n}} \quad (4.12)$$

In a metal, the index of refraction $\hat{n}$ is complex, resulting in a complex θ_t within the metal, according to Equation 4.12 . This result is non-physical. Instead, the absorption of the

incident wave should be derived using the full description of the fields within the metal. The method presented in Born and Wolf, chapter 14.1 is followed.[25] The coordinate system for this derivation is shown in Figure 4.8. For a plane wave of the form:

$$E(\omega, s) = E_0 e^{i\omega t} e^{i\hat{k}(r \cdot s)} \quad (4.13)$$

propagating in the $\hat{r}$ direction, where r is the position vector relative to the first point of incidence on the medium and $\hat{k} = \frac{\omega}{c} \hat{n} = \frac{\omega}{c} (\delta + i\kappa)$. Within the metal, the propagation vector $s^{(t)}$ is given in the x,z plane as:

$$\begin{aligned} s_x^{(t)} &= \sin(\theta_t) \\ s_y^{(t)} &= 0 \\ s_z^{(t)} &= \cos(\theta_t) \end{aligned} \quad (4.14)$$

using equations and , and defining $\hat{n} = \delta + i\kappa$ results in:

$$s_x^{(t)} = \sin(\theta_t) = \frac{\delta^2 - i\kappa}{(\delta^2 + \kappa^2)^2} \sin^2(\theta_i) \quad (4.15)$$

$$s_z^{(t)} = \cos(\theta_t) = \sqrt{1 - \sin^2(\theta_i)} = \sqrt{1 - \frac{\delta^2 - \kappa^2}{(\delta^2 + \kappa^2)^2} \sin^2(\theta_i) + i \frac{2\kappa}{(\delta^2 + \kappa^2)^2} \sin^2(\theta_i)} \quad (4.16)$$

Born and Wolf define the real quantities q and γ to describe the real and imaginary components of $s_z^{(t)}$:

Squaring equations 4.15 and 4.15, equating the real and imaginary components results in:

$$q^2 \cos(2\gamma) = 1 - \frac{\delta^2 - \kappa^2}{(\delta^2 + \kappa^2)^2} \sin^2(\theta_i) \quad (4.17)$$

$$q^2 \sin(2\gamma) = \frac{2\kappa}{(\delta^2 + \kappa^2)^2} \sin^2(\theta_i) \quad (4.18)$$

The spatial component of the incident plane wave is then given by:

$$\hat{k}(r \cdot s) = \frac{\omega}{c} (x \sin(\theta_i) + z(\delta q \cos(\gamma) - \kappa q \sin(\gamma)) + iz(q\delta \sin(\gamma) + q\kappa \cos(\gamma))) \quad (4.19)$$

From this form, the absorption length as a function of angle within the metal can be calculated. The complex part of the index of refraction is responsible for absorption within the material. The absorption length is defined as the length over which the time average energy density of the propagating wave within the medium to $\frac{1}{e}$ of its original value:

$$E(\omega, s) = E_0 e^{i\frac{\omega}{c} \kappa(r \cdot s)} \cos \left(\omega \left(\frac{n}{c} (r \cdot s) - t \right) \right) \quad (4.20)$$

The time average real part gives energy density ω :

$$w = \bar{E}^2 = \omega_0 e^{-\chi(r \cdot s)} \quad (4.21)$$

where $\chi = \frac{2\omega}{c}\kappa = \frac{4\pi}{\lambda}\kappa$ Therefore, the optical absorption length $\lambda_{opt}(\theta_i)$, is defined as:

$$\lambda_{opt}(\theta) = \frac{\lambda}{4\pi} \left(\frac{1}{q\delta \sin(\gamma) + q\kappa \cos(\gamma)} \right) \quad (4.22)$$

To solve Equation 4.22 in terms of known angles and material parameters θ_i, κ, δ , the trigonometric identities $\cos(2\gamma) = 2\cos^2(\gamma) - 1$ and $\cos(2\gamma) = 1 - 2\sin^2(\gamma)$ are used in Equation . Solving for the quantities $q \cos(\gamma)$ and $q \sin(\gamma)$:

$$q \cos(\gamma) = \sqrt{\frac{q^2 + 1}{2} - \frac{\gamma^2 - \kappa^2}{2(\delta^2 + \kappa^2)^2} \sin(\theta_i)} \quad (4.23)$$

$$q \sin(\gamma) = \sqrt{\frac{q^2 + 1}{2} + \frac{\gamma^2 - \kappa^2}{2(\delta^2 + \kappa^2)^2} \sin(\theta_i)} \quad (4.24)$$

Finally, for an expression for q , the system of equations and are solved simultaneously:

$$q = \left[\left(1 - \frac{\delta^2 - \kappa^2}{(\delta^2 + \kappa^2)^2} \sin^2(\theta_i) \right)^2 + \left(\frac{2\kappa}{(\delta^2 + \kappa^2)^2} \sin^2(\theta_i) \right)^2 \right]^{1/4} \quad (4.25)$$

The complete expression for $\lambda_{opt}(\theta_i)$ in Equation can now be evaluated.

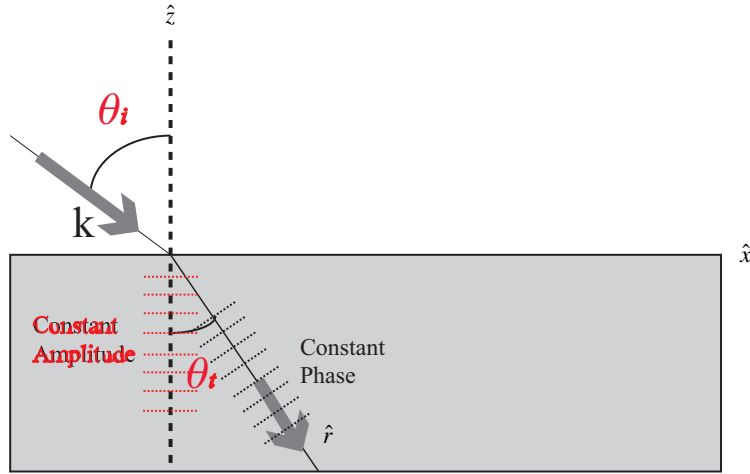


Figure 4.8: The relationship of the angle of refraction to that of planes of constant phase and constant amplitude within the metal. Planes of constant amplitude occur parallel to the surface. Planes of constant phase occur at the angle θ_t . The resulting wave within the metal is inhomogeneous.

Snell's Law for the complex index of refraction:

The dependence of the angle of refraction to that of the angle of incidence is also attained from this formalism. From Equations and , surfaces of constant phase and constant amplitude are apparent. If Equation is represented as $\hat{k}(r \cdot s) = \frac{\omega}{c}(n_{re} + in_{im})$, Equation becomes:

$$E(\omega, s) = E_0 e^{i\omega t} e^{i\hat{k}(r \cdot s)} = E_0 e^{i\omega t} e^{\frac{\omega}{c} n_{im}} e^{i\frac{\omega}{c} n_{real}} \quad (4.26)$$

Planes of constant amplitude, (4.8) for waves propagating within the metal are given by the term:

$$e^{\frac{\omega}{c} n_{im}} = e^{\frac{\omega}{c} qz(\delta \sin(\gamma) + \kappa \cos(\gamma))} \quad (4.27)$$

where z is a constant.

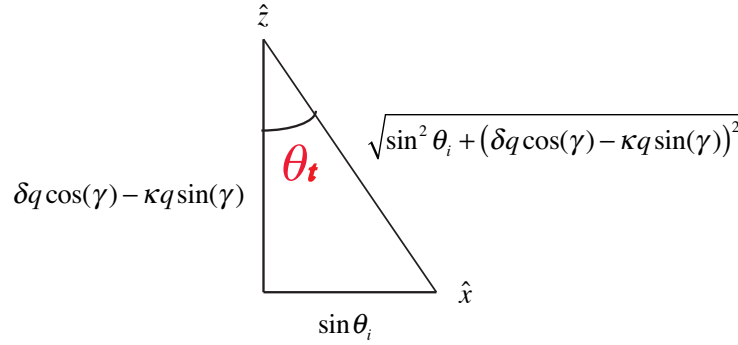


Figure 4.9: Illustrates the geometrical relationship of the planes of constant phase, from which the modified Snell's Law for the metal-vacuum interface can be derived.

Similarly, planes of constant phase occur at the refraction angle as measured from the normal to the boundary, θ_t and are given by the term:

$$e^{i\frac{\omega}{c} n_{real}} = e^{iz\frac{\omega}{c} [x \sin(\theta_i) + qz(\delta \cos(\gamma) - \kappa \sin(\gamma))]} \quad (4.28)$$

where

$$x \sin(\theta_i) + qz(\delta \cos(\gamma) - \kappa \sin(\gamma)) \quad (4.29)$$

is a constant. From the geometrical relationship (Figure 42) between θ_i and θ_t can be derived:

$$\sin(\theta_t) = \frac{\sin(\theta_i)}{\sqrt{q^2(\delta \cos(\gamma) - \kappa \sin(\gamma))^2 + \sin^2(\theta_i)}} \quad (4.30)$$

$$\cos(\theta_t) = \frac{\delta \cos(\gamma) - \kappa \sin(\gamma)}{\sqrt{q^2(\delta \cos(\gamma) - \kappa \sin(\gamma))^2 + \sin^2(\theta_i)}} \quad (4.31)$$

For n defined as

$$n' = \sqrt{q^2(\delta \cos(\gamma) - \kappa \sin(\gamma))^2 + \sin^2(\theta_i)} \quad (4.32)$$

, a form of Snells law for the case of a complex index of refraction results from Equation :

$$\sin(\theta_t) = \frac{\sin(\theta_i)}{n'} \quad (4.33)$$

Final form of the escape probability (Step 2):

Equation shows the form of the escape probability neglecting the angular contribution caused by the angle of incidence of the incoming ray. When including such effects, matching of the depth at which the electron is produced and the mean free path of the electron is required. The distance from the surface at where the electron is produced, z , is given by $z = \lambda_{opt}(\theta_i) \cos(\theta_t)$, where θ_t is the angle of refraction within the metal. The escape probability, F_{e-e} , then becomes:

$$F_{e-e} = \frac{1}{1 + \frac{\lambda_{opt}(\theta_i) \cos(\theta_t)}{\lambda_{e-e}}} \quad (4.34)$$

where $\cos(\theta_t)$ is given by Equation (4.31).

The effect of including the effect of the angle of incidence in the Three-Step Model are depicted in Figure 4.10. The Palik dataset from the DABAX data files in XOP are used for material constants at low photon energies.[26] Solid and dotted lines represent the evaluation of the model with and without the angular corrections, respectively. Quantitatively, the maximum enhancement of the quantum efficiency due to angular effects in Copper is 22% at 62 degrees. The goal of the Three-Step Model of Photoemission is to predict of the absolute Quantum Efficiency (QE) expected as a function of photon energy and angle. In order to predict the relative electron yield, however, the QE must be normalized. For the purposes of representing the electron yield, $Y(\theta_i)$, the QE is normalized to its value at normal incidence:

$$Y(\theta_i) = \frac{QE(\theta_i)}{QE(0)} \quad (4.35)$$

This relationship is used in the remainder of the document for the reporting of AREY datasets. Figure 4.11 shows AREY data taken on the (100) face of a single-crystal copper sample. The dataset were taken at photon energies ($E_{photon} - \Phi$) of 5.55 and 5.89 eV, respectively. In the figure, the solid black line is three-step model calculation results at 5.89 eV. It can be seen that the calculation results fit the dataset very well. Only a small change in only a small enhancement in yield is seen at large angles, commensurate with the predictions of the Three-step model.

Figure 4.12 is AREY data taken on the (111) face of single crystal copper. Once again, the solid black line represents the fit of the AREY dataset to the three-step model normalized yield functions. In this case, however, the fit completely fails to describe the AREY dataset.

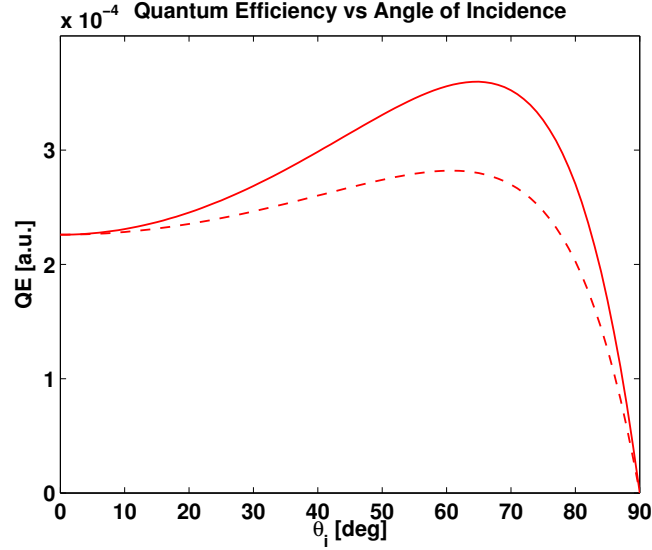


Figure 4.10: The predicted quantum efficiency as a function of angle of incidence using the Three-Step Model of Photoemission evaluated for Copper at a photon energy of 5.77 eV. Dotted lines represent the three-step model result without angle of incidence corrections. The assumed photon energy is set such that the excess energy is 0.83 eV. Zero accelerating voltage is assumed. At the maximum (62 degrees), inclusion of the angular corrections results in a 22% change in QE for copper.

In this case we see that the relative yields are close to 20 at the optimum angle, around 70 degrees angle of incidence, for p polarized light. There have been many attempts at a quantitative description of the absolute quantum efficiency of the angular enhancement based on electronic state theory.[27, 28, 29] Many of these methods introduce surface and volume forms of the index of refraction in order to reconcile the angular enhancement with the Fresnel-derived equations. Junker, Waldron and Jaccodine (JWJ) present a method of predicting the shape of the angular yield through a single fitting parameter B and a single index of refraction.[30] The key point in the JWJ theory is that it assigns extra significance to the component of E that is perpendicular to the surface, by a factor of B in electric field. The physical origin of this is not explained; it is purely a phenomenological model. JWJ speculate that the origin lies in the extreme gradient of potential at the surface, but no evidence is given. The JWJ ansatz is used to predict the shape of the AREY datasets. Using the coordinate system depicted in Figure 40, the excited photocurrent density J_z , is expressed in terms of the electric field amplitudes within the material:

$$J_z = D' (B|E_z|^2 + |E_y|^2 + |E_x|^2) g \quad (4.36)$$

Where D' is a parameter attributed to surface roughness, and g is a parameter including the effects of work function, Fermi energy, temperature and the energy of the absorbed photon.

The total current is given by $J_z A \sec(\theta)$ where $A \sec(\theta)$ represents the area of the beam and the angle of incidence, respectively. Normalizing the predicted current as a function of angle to that of the emission at normal incidence in the manner of Equation (4.38) cancels the effects of D' and g in (??), and leads to an expression for the expected photoemitted yield as a function of angle [30]:

$$Y(\theta) = \left| \frac{n+1}{n_0 - n^2 \cos(\theta)} \right|^2 \left[\frac{1+p}{2} |n_0|^2 + \frac{1-p}{2} \left| \frac{n_0 + n^2 \cos(\theta)}{n_0 + \cos(\theta)} \right|^2 + \frac{1+p}{2} C_v \sin^2(\theta) \right] \cos(\theta) \quad (4.37)$$

where n = index of refraction, $n_0 = \sqrt{n^2 + \sin^2(\theta)}$. The parameter p is constant, expressing the degree of polarization of the incoming wave, normalized by the total intensity I as:

$$p = \frac{(I_{\parallel} - I_{\perp})}{I} \quad (4.38)$$

Finally, the factor C_v defines the effect of the B parameter weighted by the index of refraction inside of the material:

$$C_v = |n|^4 B = (\delta^2 + \beta^2)^2 B \quad (4.39)$$

Figure 4.12 shows the fit of the JWJ Equation (4.37) to the AREY data taken for Cu(111) dataset. The figure shows good agreement of the data to the JWJ ansatz for differing B values.

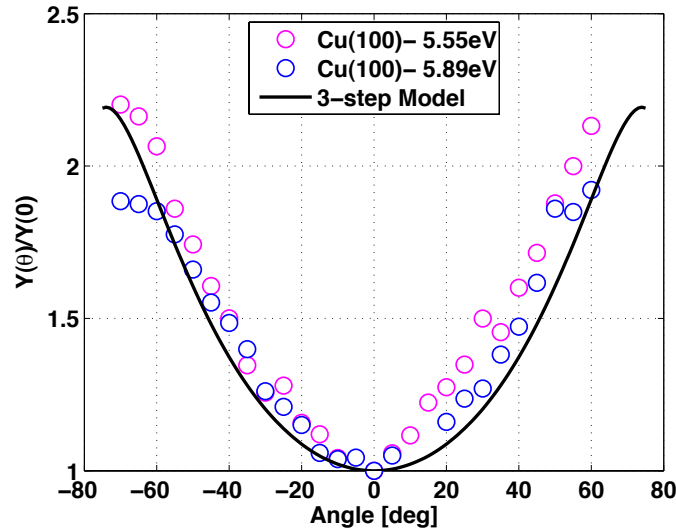


Figure 4.11: Prediction of the AREY for Cu(100) at photon energies of 5.55 and 5.89 eV. The modified form of the Fresnel equations are fit at 5.89 eV to produce the solid black line. A single parameter, θ_0 , the angle offset is fit.

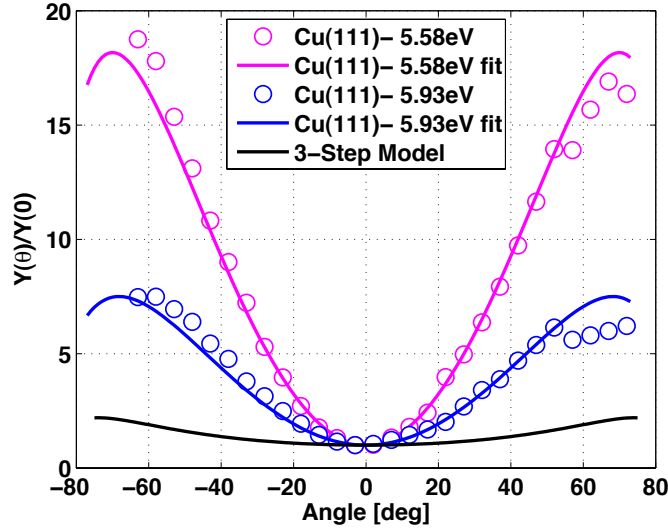


Figure 4.12: Prediction of the AREY for Cu(111) at photon energies of 5.58 eV and 5.93 eV. The solid black line fit represents the AREY as predicted by the Fresnel equations. The data are fit using the angular enhancement as predicted by the Vectorial Photoelectric Effect. Two fit parameters are used for each dataset: B , the enhancement factor of the z-component of the Electric field, and θ_0 , the angular offset.

4.5 Single Crystal Studies: Determination of Electronic Structure by Angle Resolved Photoemission (ARPES)

The final evaluation technique involves the determination of the photocathode electronic structure. As described in Chapter 3, the Time-of-Flight (TOF) method is used to measure the momenta of the photoemitted electrons. This section begins with a description of the methods used to calibrate and characterize the contact potential and sample distance in the parallel plate detector configuration. Equations for calculation of the electron energy and momenta leaving the photocathode are then discussed. Finally, angle integrated and angle resolved datasets taken from a clean Cu(111) photocathode are presented.

Determination of Contact Potential

Figure 4.13 depicts the experimental geometry under consideration. The sample under test forms a parallel plate capacitor at a distance d from the entrance grid of the detector. The grid is kept at ground potential and the bias V_{bias} is placed on the sample to provide an accelerating or decelerating potential. The emitted photoelectrons follow a parabolic path

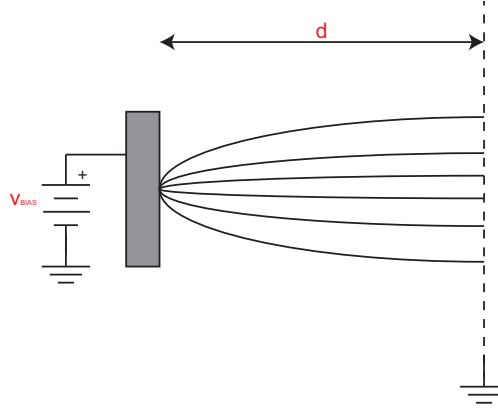


Figure 4.13: The parallel plate detector geometry and potentials. Photoelectrons follow the parabolic trajectory once emitted from the cathode. Relevant parameters are the sample-detector distance d , and the imposed sample bias V_{bias} .

from the sample to the detector, as shown.

With $V_{bias} = 0$, there exists a small potential between the sample and detector due to the differences in the work functions of the two materials, namely the Contact Potential, ϕ_{CP} .^[31, 32] In order to calibrate the sample-detector system for this potential, the number of electron counts as a function of an imposed retarding potential on the sample is measured. Knowledge of the work function and the Fermi energy of the sample are used to determine ϕ_{CP} . (Figure 4.14)

The retarding field method determines the voltage at which the highest kinetic energy electrons can no longer reach the detector. This voltage, less the maximum initial kinetic energy of the electron, is ϕ_{CP} . Figure 4.15 shows the measured electron intensity as a function of retarding potential. The retarding field method was used with a cathode of clean Cu(111) and a photon energy of 5.77eV. The maximum kinetic energy of the photoelectrons occurs at $E_{photon} - E_F = 0.82\text{eV}$. The collected dataset cutoff is 2.7eV, representing the sum of the maximum electron kinetic energy and the detector contact potential. The difference, ϕ_{CP} , is therefore 1.88eV, and represents an inherent accelerating potential between sample and detector for the Cu(111) system.

Calculation of the photoemitted electron energy and momenta

In Chapter 3, the TOF detector and experimental geometry were described. For each electron, the TOF detector records the position (x,y) and time (t) of the incident electron. In order to reconstruct the energy and momenta of the electron leaving the photocathode, the TOF detector data must be corrected for acceleration due to the imposed bias voltage.

Under the assumption of the parallel plate geometry, the field lines between sample and detector are assumed to be parallel. Therefore, electron acceleration due to the imposed bias

(V_{bias}) occurs solely in the $\hat{z}$ direction. For an accelerating bias of

$$V_{eff} = V_{bias} + V_{CP} \quad (4.40)$$

, the components of the electron velocity at the sample surface are given by:

$$\begin{aligned} v_x^0 &= \frac{x-x_0}{t-t_0} \\ v_y^0 &= \frac{y-y_0}{t-t_0} \\ v_z^0 &= \frac{d}{t-t_0} - \frac{V_{eff}}{2m_e d} (t - t_0) \end{aligned} \quad (4.41)$$

where (x_0, y_0) is the coordinate of the center of mass of the emission on the detector. Time t_0 is the trigger time delay of the laser on the photocathode. This can be estimated by reflecting the UV laser into the detector and recording the arrival time channel of secondary electrons generated on the surface of the detector MCP. The x and y components of the photoelectron velocity at the photocathode determine the initial transverse momenta:

$$\begin{aligned} k_x &= \frac{m}{\hbar} (v_x^0)^2 \\ k_y &= \frac{m}{\hbar} (v_y^0)^2 \end{aligned} \quad (4.42)$$

The total energy of the photoemitted electron is given by:

$$E_{tot} = E_x^0 + E_y^0 + E_z^0 \quad (4.43)$$

where:

$$\begin{aligned} E_x^0 &= \frac{\hbar^2}{2m_e} k_x^2 \\ E_y^0 &= \frac{\hbar^2}{2m_e} k_y^2 \\ E_z^0 &= \frac{m_e}{2} (v_z^0)^2 \end{aligned} \quad (4.44)$$

The complete photoelectron energy and momenta are now determined. The momentum resolution of the system is given by: [18]

$$\Delta k_{\parallel} \approx k_{\parallel} \sqrt{\left(\frac{\sqrt{x^2 + y^2}}{d}\right)^2 + \frac{2E}{m_e} \left(\frac{\Delta t}{d}\right)^2} \quad (4.45)$$

and the energy resolution is given by:

$$\Delta E_{TOF} = \sqrt{\frac{8E_{kin}^3}{m_e} \frac{\Delta t}{d}} \quad (4.46)$$

for an initial total kinetic energy of $E = 1\text{eV}$, equation predicts

$$\Delta k_x, \Delta k_y = 2 \cdot 10^{-3} \frac{1}{\text{\AA}} \quad (4.47)$$

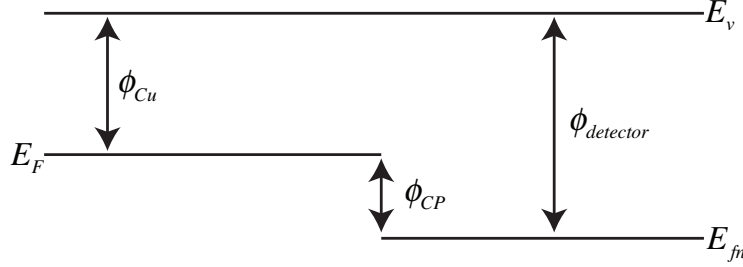


Figure 4.14: The relationship of the sample and detector work functions in defining ϕ_{CP} , the sample-detector contact potential.

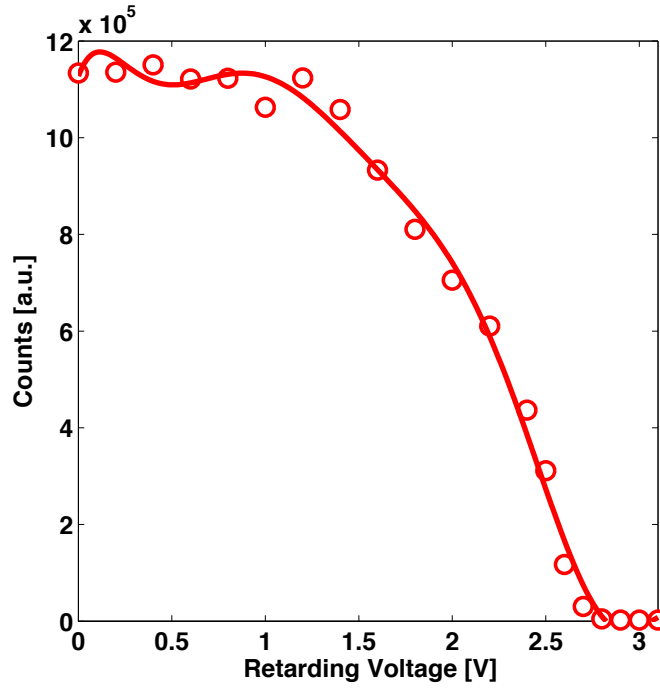


Figure 4.15: The retarding field experiment for the determination of the Contact Potential, ϕ_{CP} . The number of counts reaching the detector as a function of the retarding voltage is measured. For Cu(111) this occurs at $V = 2.7V$. Subtracting the maximum initial kinetic energy of the electrons ($E_{photon} - E_F = 0.82eV$), ϕ_{CP} is determined to be $1.88eV$. The solid line is a polynomial fit to the collected dataset and serves to guide the eye.

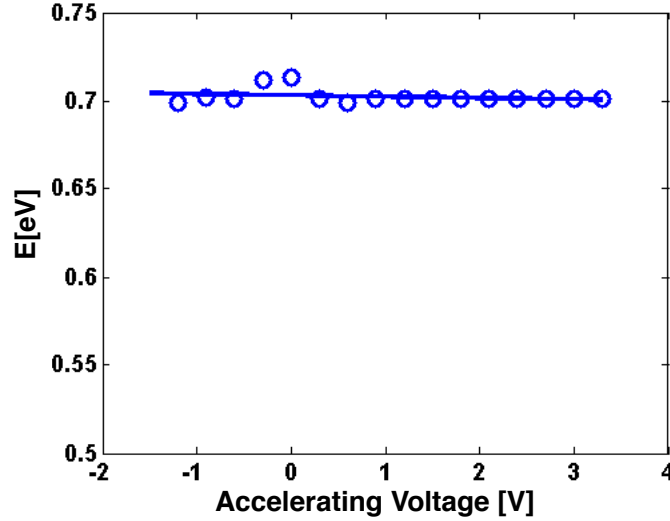


Figure 4.16: Calibration of the sample-detector distance. The energy channel of the peak number of counts is plotted as a function of the bias voltage. At the correct value of d , the voltage correction in the z -component of Equation directly cancels the applied voltage. The result is a horizontal line. For this figure, $d = 30\text{mm}$.

and an $\Delta E_{TOF} = 4.80\text{meV}$.

The final calibration step for the parallel-plate detector geometry shown in Figure 4.13 is to accurately determine the sample distance, d , by using the applied bias V_{bias} . The initial measurement of d is given by measurement of the chamber-to-detector distance, however it varies due to variation of the manipulator position and photon incident angle. The total electron energy can be expressed as:

$$E = E_x + E_y + \left(\frac{m_e}{2}\right) \frac{d}{t - t_0} - \frac{V_{\text{eff}}}{4d} (t - t_0) \quad (4.48)$$

In this form, the relationship between the bias voltage and the distance is clearly visible. The correct d is that which balances the fourth correction term with reduction in due to acceleration of the third term. An accelerating bias is therefore completely cancelled in the analysis and the E versus V_{applied} plot is a horizontal line. Figure 4.16 the measured relation of electron energies with applied voltages, for a distance $d = 30\text{mm}$.

The resulting three-dimensional dataset (E , k_x , k_y) completely describes the accessible reciprocal space (energy, momentum) of the material under test. The normal emission spectrum of Cu(111) (counts, E) is shown in Figure 4.17. The Fermi energy, E_f is clearly visible as the histogram cutoff and the width of the spectrum is proportional to the maximum energy level able to be accessed through this method ($\hbar\omega - \phi_{\text{eff}}$). The peak in the number of counts is easily identified as the Cu(111) surface state.[33, 34, 35] Figure 4.18 shows the

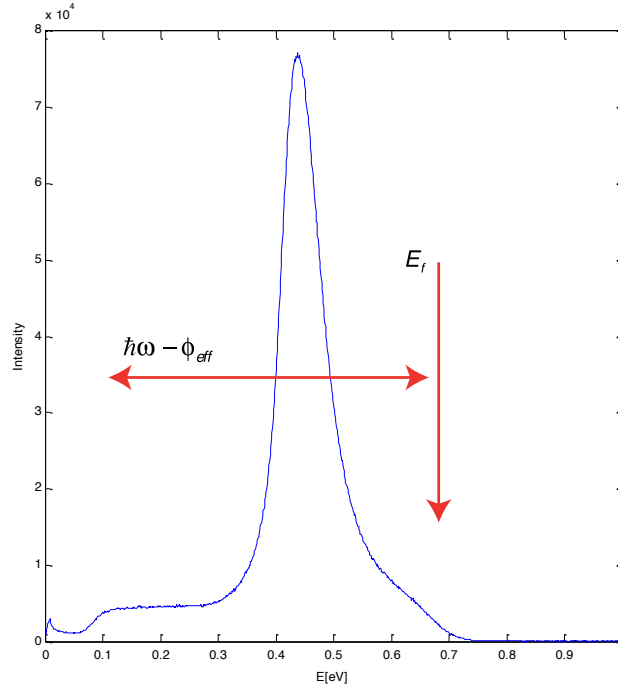


Figure 4.17: The normal emission of a Cu(111) ARPES dataset; Intensity vs. total Energy. Electron emission occurs from the lowest energy state $\hbar\omega - \phi_{eff}$ to the Fermi level, E_f .

same copper dataset expressed in an angle-resolved manner ($E, k_y, k_x = 0$). The Fermi energy and spectrum width are labeled. Figure 4.19 depicts the angle resolved dataset ($k_y, k_x, E = E_f$), relating only the transverse momentum of the photoemitted electrons in the x and y directions.

This section has described the development of a Time-of-Flight spectrometer for the measurement of low energy electrons. This spectrometer has energy resolution far less than k_{BT} , the thermal energy of electrons within the material. The parallel-plate configuration provides solely longitudinal accelerating fields that can be corrected by the application of a bias voltage on the sample. A method of correcting the sample distance has been shown to be effective in compensating the effect of an applied accelerating bias. Furthermore, the ARPES spectrum of Cu(111) has been successfully recorded. This three-dimensional dataset shows the parabolic energy momentum relationship of the Cu(111) surface state in Energy versus transverse momentum (Figure 4.18) as well as localization in transverse momentum in Figure 4.19. The ARPES data presented contradicts a fundamental prediction of the Three-Step Model presented in Chapter 2: *The production of near work function photoelectrons from the surface state is not continuous, but is truncated at a maximum emission angle.* The photoelectrons, rather, maintain the energy and momentum relationship of the state from

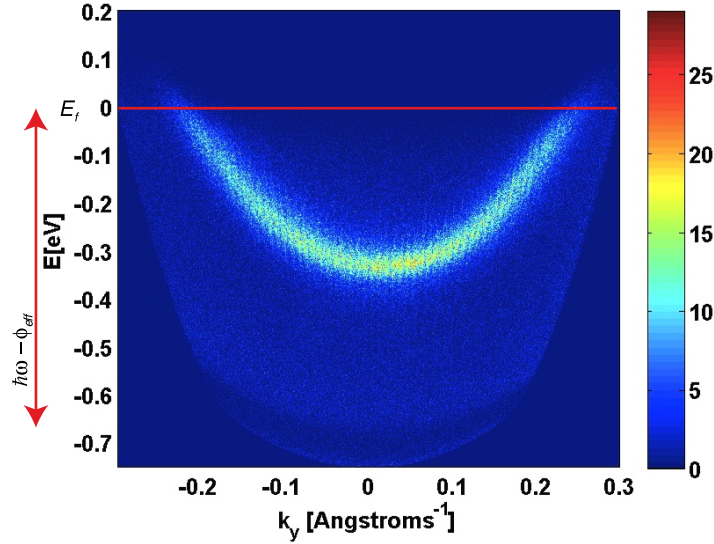


Figure 4.18: The angle-resolved spectrum of the dataset in Figure 4.16. This figure displays the total electron energy vs transverse momentum in the y-direction with zero transverse x-momentum. The colorbar represents the number of counts in each channel.

which they were excited, as is evidenced by the ARPES spectrum. The emittance of the electron beam produced with non-isotropic emission, therefore, follows the electronic structure as predicted by the energy and momentum relationship of ARPES measurements.[3, 36] Furthermore, the presence of the surface state in the Cu(111) spectrum at energies near the Fermi level is a salient feature in the electronic structure that distinguishes Cu(111) from Cu(100).[37] One could also hypothesize that the angular enhancement of the electron yield (Section 4.5) could be correlated to the presence or absence of the surface state, based on electronic structure of photocathode materials. It is well known that the emittance electron beams comprised of photoemitted electrons is solely determined by the electron transverse momentum. Therefore photoemitted electrons from the surface state of Cu(111) produce electron beams with small emittance since it has a transverse momentum cut-off due to non-isotropic emission. The experiment described in the section to follow aims to investigate these hypotheses.

4.6 Use as a Photocathode: Electron Yield in Ion-Bombarded Metal Surfaces

The previous sections of this chapter have presented data from and analysis of a suite of tools with the aim of characterizing and validating the Three-Step model of photoemission

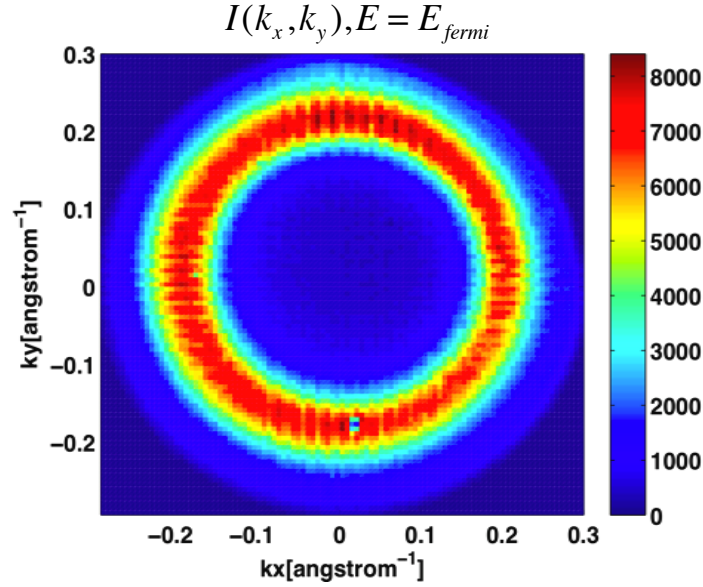


Figure 4.19: This is another visualization of the ARPES spectrum of Figure 4.16. In this figure, the transverse momentum in the y-direction is plotted against the transverse momentum in the x-direction for total energy fixed at the Fermi level, E_f . The colorbar represents the number of counts in each channel.

for metal photocathodes. The work function, AREY and ARPES studies presented have been shown to be robust and effective methods for characterizing the surface order and electronic state of metal photocathodes. In this section, these tools are used to evaluate correlations between surface order, AREY and electronic structure. This section begins with a brief discussion of the origin of the surface state. Following this, the effect of ion damage on Cu(111) surface state and AREY is discussed.

The Origins of the Surface State

First presented in the works of Tamm and Shockley, the surface state is a quasi-two dimensional state that exists due to the abrupt termination of the three-dimensional crystalline lattice.[38, 39] Under the tight-binding model of atomic potentials depicted in Figure 4.20, the surface states are represented by solutions with complex wavevectors. Figure 4.21 elucidates the bulk state wavefunction and the surface state wavefunction. First, the surface state is localized to the two-dimensional surface, as elucidated by the decay of the wavefunction into the crystal. Secondly, the surface state represents a net buildup of charge at the surface boundary; the characteristics of which depend on the interatomic spacing and adjacency of electron band crossings in the material system of interest. The ion damage ansatz is the following: if the crystal-vacuum interface of the material is interrupted such

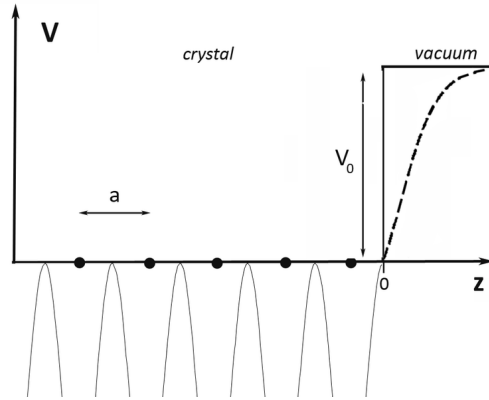
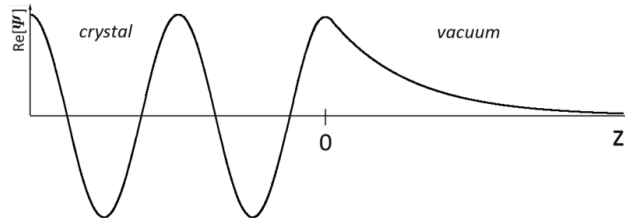
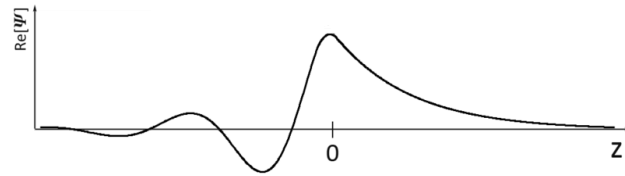


Figure 4.20: The crystal-vacuum interface under the Tight Binding atomic potential approximation. [19]



(a) Bulk State Wavefunction solutions



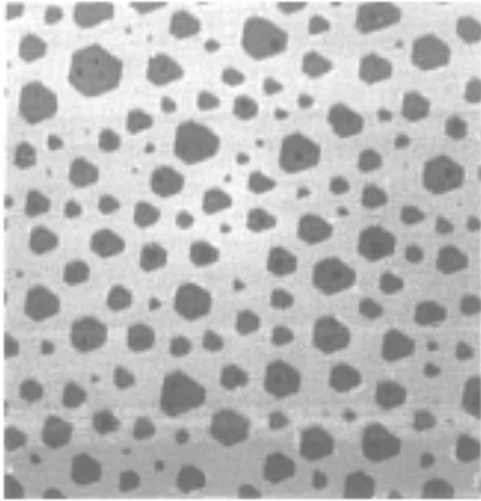
(b) Surface State Wavefunction solutions

Figure 4.21: Plots of the wavefunction solutions under the potential assumed in Figure 4.19. The surface state is clearly shown to be localized at the surface in 4.21b as it decays both in the crystal and vacuum.[19]

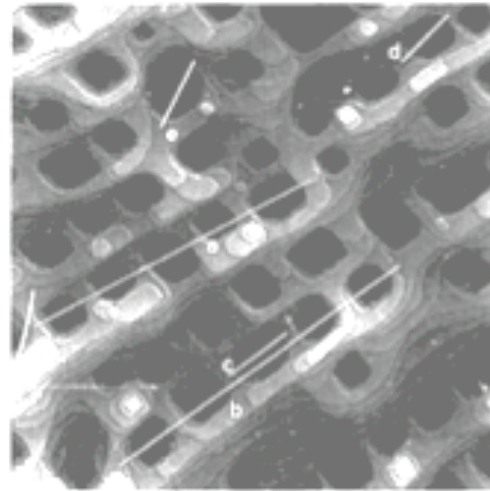
that its lateral scale cannot support a surface state, does an observable reduction in the AREY and a change in the corresponding electronic structure occur? This is the subject of the next section.

4.7 Ion damage studies of Cu(111)

The effect of ion damage on crystalline metallic surfaces has been well studied.[40, 41, 42] This is of particular interest in the case of photocathodes for RF electron guns, as the accelerating gradient causes ionized gas products to ablate the surface of the photocathode. Figure 4.22 shows SEM photographs of ion bombarded Pt(111) and Cu(001) surfaces under light (a) and heavy (b) Ar⁺ ion bombarding. The result is the creation of small vacancy islands of the order of 5–80 nm, the density of which increases with increasing ion dose.[41]



(a) An SEM photo ($825 \times 825 \text{ \AA}^2$) of ion bombarded Pt(111) under light damage (Fluence: $3.45 \times 10^{16} \text{ ions/cm}^2$).



(b) The effect of high damage (Fluence: $5.4 \times 10^{15} \text{ ions/cm}^2$ at 45 Deg angle of incidence), SEM photo ($200 \times 200 \text{ nm}^2$) of ion bombarded Cu(001).

Figure 4.22: The effects of Ar⁺ ion bombardment on metallic surfaces.[41, 42]

In this experiment, a clean Cu(111) photocathode underwent a series of steps of Ar⁺ ion-bombarding. (2keV, 15 min exposure) At each step, work function, AREY and ARPES measurements were conducted. Figure 4.23 shows the measured work function as a function of ion damage. The dataset shows an initial sample in agreement with published values.[43] This value is reduced as the sample is exposed to further ion damage until it reaches a minimum at 4.38eV. Between steps 6 and 7, the sample is annealed and the work function returns to 4.88 eV. Similarly, in Figure 4.24, the AREY is shown as a function of angle for each of the work functions in Figure 4.23. The AREY trend follows that of the work function;

as the work function decreases, the measured AREY values decrease correspondingly to a minimum that matches the Fresnel-derived AREY. When the sample is annealed, the large enhancement is once again observed. Finally, Figure 4.25 shows ARPES measurements taken at successive steps in the ion damage experiment. In panel 1, the surface state is clearly visible. As the ion bombardment process continues, the surface state intensity diminishes until panel 6, where it is no longer observable. After an anneal step, panel 8 shows the return of the surface state. Our results show that the increase of photoemitted electron yield at grazing incidence angle using P-polarization light is completely from the surface state of Cu(111). Furthermore, the presence or absence surface state is controllable using ion-bombarding and annealing cycle.

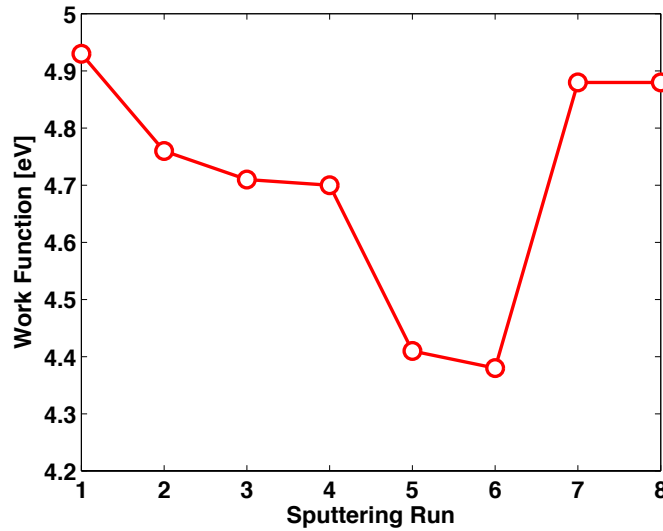


Figure 4.23: The measured workfunction as a function of ion damage for a Cu(111) sample. Each ion damage step is comprised of bombardment of 2keV Ar+ ions for 15 minutes. Between steps 6 and 7, the sample is annealed at 500C for 2 hours. The effect of ion damage to the surface is a lowering of the observed workfunction. This damage is trivially removed by annealing the sample.

4.8 Use as a Photocathode: Calculation of the Transverse Emittance from the ARPES spectrum

Chapter 2 defines the normalized emittance as:

$$\varepsilon_n = \sigma_x \sigma_{p_x} \quad (4.49)$$

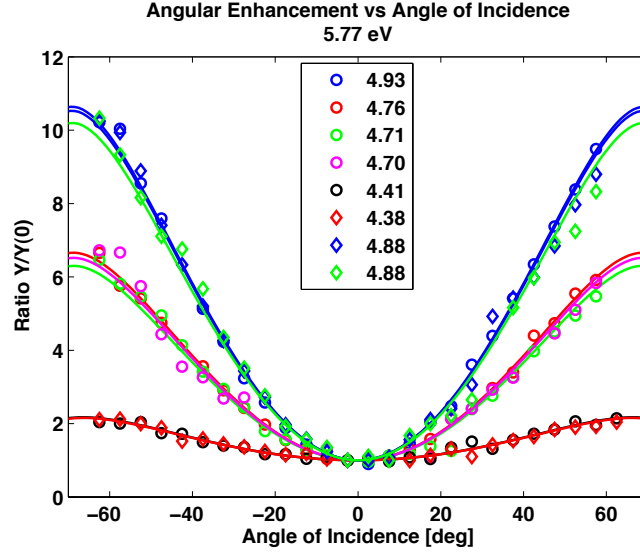


Figure 4.24: AREY data for each of the measured work functions, fit with the JWJ prediction (Equation (4.36)). The angular enhancement decreases with increasing damage, following a similar trend of the surface work function in Figure 4.23. At the minimum enhancement, the data is well fit with the angular prediction of the Fresnel equations as described by Equation (4.37).

where σ_x is the root-mean-square beam size [mm] and σ_{p_x} , the transverse momentum is given by:

$$\sigma_{p_x} = \frac{\sqrt{\langle p_x^2 \rangle}}{mc} \quad (4.50)$$

The individual particle momentum as measured by the Time of Flight method can be expressed as:

$$p_{\parallel} = \hbar \sqrt{k_x^2 + k_y^2} \quad (4.51)$$

leading to the definition of the RMS normalized emittance for a 1 mm beam:

$$\varepsilon_{RMS} = \frac{1}{2} \frac{\hbar k_{max}}{mc} \quad (4.52)$$

It can be seen from Figure 4.18 that the maximum transverse momentum from the Cu(111) surface state is 0.251 Angstroms, yielding an emittance of 0.4 um mrad/mm (rms). At the limiting case of the Fermi level, the actual emittance is significantly lower due to averaging over the entirety of the surface state band.

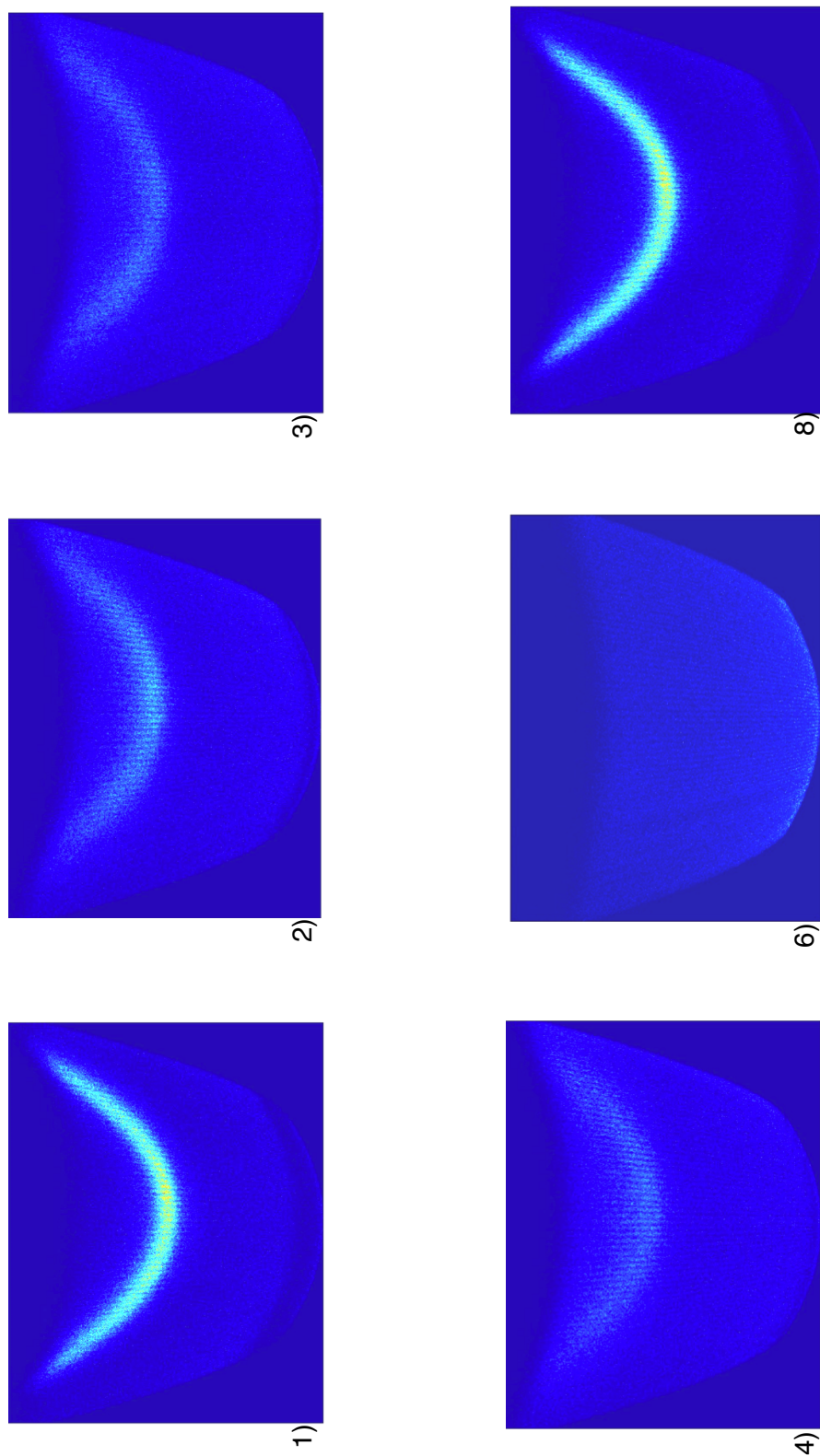


Figure 4.25: The electronic structure observed trend in the surface state intensity as a function of ion damage. The pictures are labeled with their corresponding sample numbers, as shown in Figure 4.23. The trend indicates a reduction in surface state emission intensity which correlates with the lowering of the sample work function.

4.9 Summary

This chapter presented the analysis methods for analyzing the AREY, ARPES and work function for metal photocathodes for free electron laser applications. A copper photocathode from the Stanford LCLS was analyzed and found to be comprised of large polycrystalline grains. This discovery lead to an attempt to differentiate the roles of the predominant single crystalline faces of Copper (Cu(100) and Cu(111)) present on the face of the photocathode. Through measurement of the work function, AREY and ARPES the role Cu(111) surface state was identified as a cause of the differing AREY behavior of the material orientations. In addition, a reduction in thermal emittance of the related beam was measured, also correlated to the presence of the surface state.

In terms of potential applications, this effect is extremely valuable: ordered surfaces of the correct orientation give yield a factor of 20 higher than disordered materials corresponding to a reduction in average transverse momentum. It also leads to a way to improve the effect. For example, Ag(111) also supports a surface state, but in this case it is far narrower (10 meV) and far closer to the Fermi level (60 meV) than in the Cu(111) case. The cut of the surface state at the Fermi level is roughly 2.5 x smaller than in the Cu case, resulting in lower transverse emittance. It may perhaps give a greater relative yield, due to the high density of states in the intrinsically narrower state. It also will lead us to look for similar band effects in bulk band structures, such as in direct bandgap semiconductors and in more complex materials with surface states that cross at the Fermi level at zero $k_{x,y}$, namely the recently discovered topological insulators.

Chapter 5

Summary and Conclusions

5.1 Summary of Results

This work has described a cathode laboratory for the validation and characterization of metal photocathodes for Free Electron Laser applications. In the laboratory, A UHV chamber with 1×10^{-10} torr vacuum and magnetic field shielding as fabricated and is demonstrated to be capable of studying photoemitted low energy electron close to the Fermi level of the photocathode. A five dimensional manipulator is used to fully control the geometry of the sample and can be used to measure the angular dependence of emitted electrons. The surface properties of the photocathode materials is controlled using ion-bombarding and annealing techniques and is characterized by LEED method. A wavelength tunable CW plasma laser is used to measure the quantum efficiency of the photocathode. The ARPES measurement is done by using a delay-line-detector and a Ti:Sapphire oscillator with picosecond duration. The laboratory is equipped with advanced control software systems for baking, pumping, sample rotating and data acquisition based on Labview code. The aim of experiments conducted herein has been to determine the validity of the Three-Step Model of photoemission in predicting the total quantum yield and Quantum Efficiency of photoemissive materials. In particular, four major conclusions can be drawn from the work presented here:

1. Near-threshold emission from polycrystalline photocathodes is dominated by the behavior of single-crystalline grains; in the limit that the photon beam is smaller than the emissive grain size.
2. Up to a factor of 20 enhancement of quantum yield is observed than that predicted by the Fresnel Equations. This enhancement has been shown to be correlated to sample surface order and electronic structure; the enhancement occurs on surfaces supporting a surface state.
3. Surface state emission yields a factor in 3 reduction in beam emittance than that predicted for the Three Step Model for Cu(111) through the limitation of the transverse momentum of electrons comprising the photoemissive beam.

4. Enhancement and emittance reduction is correlated to surface order, which can be trivially restored from the effects of ion damage thorough renewal of the surface order by thermal annealing cycles to recover the sample workfunction.

These results could have great impact on the selection and engineering of photoemissive cathodes for Free Electron Lasers (FELs). Photoemission from the electron band states within the material and reduction in the Fermi energy can be used as tools for achieving high quantum yield and beam emittance reduction. Material systems could then be chosen such that the energy-momentum relationship of the surface state occurs closer to the Fermi Energy. Silver (111), for example, is one such candidate system, in which the surface state occurs 0.28 eV closer to the Fermi Energy than that of Copper (111). According to the model presented here, a further factor of 3 reduction in emittance could be obtained by emission using Ag(111) as the photocathode.

In terms of potential applications, the use of ordered surfaces as photocathodes is extremely valuable; not only do surfaces of the correct orientation gives a factor of 20 higher than disordered materials, the enhanced yield also corresponds to reduced average transverse momentum. Further enhancement is also possible. For example, Ag(111) also supports a surface state, but in this case it is far narrower (10 meV) and far closer to the Fermi level (60 meV) than in the Cu(111) case. The cut of the surface state at the Fermi level is roughly 2.5x smaller than in the Cu case, resulting in lower transverse emittance. It may perhaps give a greater relative yield, due to the high density of states in the intrinsically narrower state. It also will lead us to look for similar band effects in bulk band structures, such as in direct bandgap semiconductors and in more complex materials with surface states that cross at the Fermi level at zero $k_{x,y}$, namely the recently discovered topological insulators.

In summary, before this work there was a belief that photocathodes made of polycrystalline materials were optimum. The conclusion of this work is that this is not true, and huge advantages can be gained by use of ordered materials. An extension of this work will look at how materials can be engineered, for example by Molecular Beam Epitaxy, to give the desired properties of very high quantum efficiency and very low transverse momentum.

The impact of this result directly influences the scale, cost and operating range of FEL system designs in the future. For a given specification of machine parameters, either smaller, lower-cost, accelerators can be employed for a fixed range of wavelengths or a machine could be built to take advantage of a wider range of achievable photon energies for a given machine scale.

Appendix A

MATLAB Source Code

This section lists Matlab source codes used in the analysis of the data presented in this thesis.

A.1 Photoemission as a function of Angle of Refraction (aoi_refraction_angle.m)

```
%Cu(111)
work_function = 4.94;%eV
E_fermi = 7; %eV
extraction_field = 0; %[MV/m]
material_file = '/Users/greaves/Documents/Research/Photoemission/Photoemission/↵
Documents/Analysis/3_step_model/materials/palik-Cu.txt';
plot_all = 0;
lambda = 1239.842/5.77;
aoi = (0:1:90)*pi/180;

[QE_cu,s1,s2,s3,enorm,E_Excess]= spicer_3_step(work_function,E_fermi,↵
extraction_field,lambda,aoi,material_file,plot_all);
[QE_c,s1_c,s2_c,s3_c,theta_t,lambda_opt]= lsbl_calculations(work_function,E_fermi,↵
extraction_field,lambda,aoi,material_file,plot_all);

%Ag(111)
work_function = 4.74;%eV
E_fermi = 5.5; %eV
extraction_field = 0; %[MV/m]
material_file = '/Users/greaves/Documents/Research/Photoemission/Photoemission/↵
Documents/Analysis/3_step_model/materials/palik-Ag.txt';
plot_all = 0;
lambda = 1239.842/5.57;
aoi = (0:1:90)*pi/180;

[QE_ag,s1,s2,s3,enorm,E_Excess]= spicer_3_step(work_function,E_fermi,↵
extraction_field,lambda,aoi,material_file,plot_all);
```

```

[QE_c_ag,s1_c_ag,s2_c_ag,s3_c_ag,theta_t_ag,lambda_opt_ag]= lsbl_calculations(↵
    work_function,E_fermi,extraction_field,lambda,aoi,material_file,plot_all);

figure(1)
plot(aoi*180/pi,s2_c_ag,'b-',aoi*180/pi,s2_c,'r-','LineWidth',1.1);
set(gca,'fontsize',13,'fontweight','b');
legend('Ag- 5.57eV','Cu- 5.77eV','Location','Best');
xlabel('\theta_{i} [deg]');
ylabel('F_{e-e}');
title('Escape Probability vs Angle of Incidence');
set(gca,'Xlim',[0,90]);

figure(2)
plot(aoi*180/pi,QE_c_ag,'b-','LineWidth',1.1);
hold on
plot(aoi*180/pi,QE_ag,'b-','LineWidth',1.1,'HandleVisibility','off');
plot(aoi*180/pi,QE_c,'r-','LineWidth',1.1);
plot(aoi*180/pi,QE_cu,'r-','LineWidth',1.1,'HandleVisibility','off');

set(gca,'fontsize',13,'fontweight','b');
legend('Ag- 5.57eV','Cu- 5.77eV','Location','Southwest');
xlabel('\theta_{i} [deg]');
ylabel('QE [a.u.]');
title({'Quantum Efficiency vs Angle of Incidence'});
set(gca,'Xlim',[0,90]);
set(gca,'Ylim',[0,3.7e-4]);

figure(10);
plot(aoi*180/pi,lambda_opt_ag,'b-',aoi*180/pi,lambda_opt,'r-','LineWidth',1.1);
legend('Ag- 5.57eV','Cu- 5.77eV','Location','Best');
set(gca,'fontsize',13,'fontweight','b');
xlabel('\theta_{i}[deg]');
ylabel('\lambda_{opt} [nm]');
title('Optical Absorption Length vs Angle of Incidence');
set(gca,'Xlim',[0,90]);

figure(11);
plot(aoi*180/pi,theta_t_ag*180/pi,'b-',aoi*180/pi,theta_t*180/pi,'r-','LineWidth'↵
    ,1.1);
legend('Ag- 5.57eV','Cu- 5.77eV','Location','Best');
set(gca,'fontsize',13,'fontweight','b');
xlabel('\theta_{i} [deg]');
ylabel('\theta_{t}[deg]');
title({'Angle of Refraction vs Angle of Incidence'});
set(gca,'Xlim',[0,90]);

figure(12)
plot(aoi*180/pi,s1_c_ag,'b-',aoi*180/pi,s1_c,'r-','LineWidth',1.1);
set(gca,'fontsize',13,'fontweight','b');
legend('Ag- 5.57eV','Cu- 5.77eV','Location','Best');
xlabel('\theta_{i} [deg]');
ylabel('R [a.u.]');
title({'Reflectivity vs Angle of Incidence'});
set(gca,'Xlim',[0,90]);

```

A.2 Three Step Model Calculations (lsbl_calculations.m)

```

%Calculates the Quantum Efficiency of a metal using Spicer's 3 step model
%Using Dave Dowell's formulation

function [QE,R2,Fee,E_pe,theta_t,lambda_opt]= lsbl_calculations(work_function,↵
    E_fermi,extraction_field,lambda,aoi,material_file,plot_all)

%Calculate Shotkey lowering of the barrier
phi_schottky = 0.037947*sqrt(extraction_field); %eV

%Material data file
indexfile = textread(material_file,'','headerlines',1);
file_lambda = indexfile(:,1)./10; %um
file_delta = indexfile(:,2);
file_beta = indexfile(:,3);

%Choose just the index of refraction corresponding to the lambda
%range selected
delta = interp1q(file_lambda,file_delta,lambda)';
beta = interp1q(file_lambda,file_beta,lambda)';

E = 1239.842./lambda; %eV
n = delta+1i.*beta

phi_eff = work_function - phi_schottky;

%Constants for calculation of the e-e scattering crossection
lambda_m = 2.2; %um
Em = 8.6;%eV
ee_photon_energy = 4.86;%eV (255nm)

%Material Reflectivity at Normal Incidence
R = (abs((1-n)./(1+n))).^2;
%Material Reflectivity at Angle of Incidence
R2 = 1- Fresnel_T_Coeff(delta,beta,aoi);

if(plot_all)
    figure(1);
    plot(lambda,R,'bo-',lambda,R2,'ro-');
    xlabel('lambda [nm]');
    ylabel('R');
    title({'Reflectivity vs Wavelength','Normal Incidence vs angle'});
    legend('Normal incidence','AOI');
    %set(gca,'Ylim',[.32,.40]);
end

%Electron-Electron scattering form check (Dowell Fig 6)
E_prime = 4:.1:10;
lambda_ee = lambda_m.*(Em./(E_prime)).^(3/2);
%Dowell Results
E_measured = [6.109,6.715,8.914,9.311];

```

```

lambda_measured = [39.167,29.333,20.333,22.667]./10;%nm
if(1)%(plot_all)
    figure(14)
    plot(E_prime,lambda_ee,'r-',E_measured,lambda_measured,'bo');
    xlabel('E-E-{fermi} [eV]');
    ylabel('e-e crossection [nm]');
    title({'\lambda_{e-e} vs Excess Energy above the Fermi Level','Dowell Fig. 6'↵
    });
end

%Optical Absorption Length
kappa = beta./delta;

qsqr = sqrt((1-(1-kappa.^2)./((1+kappa.^2).^2*delta^2)*(sin(aoi)).^2).^2 + (2*↵
    kappa*(sin(aoi)).^2)./(delta^2*(1+kappa.^2).^2)).^2);
q_cos_gamma = sqrt(qsqr/2 + 1/2 - (1-kappa.^2)./(2*(1+kappa.^2).^2*delta^2)*(sin(↵
    ao_i)).^2);
q_sin_gamma = sqrt(qsqr/2 - 1/2 + (1-kappa.^2)./(2*(1+kappa.^2).^2*delta^2)*(sin(↵
    ao_i)).^2);

n_prime = sqrt((delta.*q_cos_gamma-kappa.*q_sin_gamma).^2+sin(aoi).^2);
theta_t = asin(sin(aoi)./n_prime);

beta_corrected = 1*(beta*q_cos_gamma + delta*q_sin_gamma);
lambda_opt = lambda./(4*pi*beta_corrected);

if(plot_all)
    figure(3)
    plot(lambda,lambda_opt);
    xlabel('lambda [nm]');
    ylabel('\lambda_{opt}');
    title('\lambda_{opt} vs wavelength');
end

%Electron-Electron Scattering Length
lambda_ee_1 = 1./(1 + sqrt(phi_eff./E));
lambda_ee_2 = (2*lambda_m*(Em)^(3/2))./(sqrt(phi_eff).*E);
lambda_ee_avg = lambda_ee_1.*lambda_ee_2;
if(plot_all)
    figure(4)
    plot(E,lambda_ee_avg);
    xlabel('E [ev]');
    ylabel('\lambda_{e-e} [nm]');
    title('\lambda_{e-e} vs E');
end

%Fraction of electrons reaching the surface
Fee = 1./(1+(lambda_opt.*(cos(theta_t))./(1.*lambda_ee_avg)));

if(plot_all)
    figure(5)
    plot(E,Fee,'bo-',E,Fee,'ro-');
    xlabel('E [eV]');
    ylabel('F_{e-e} [a.u]');
    title('Probability that an electron reaches the surface vs E');
end

%Electrons available for Photoemission by DOS

```

```

E_pe = ((E_fermi + E)./(2*E)).*(1+((E_fermi+phi_eff)./(E_fermi+E))-2*sqrt((E_fermi+phi_eff)./(E_fermi+E)));
if(plot_all)
    figure(6)
    plot(E,E_pe);
    xlabel('E [eV] ');
    ylabel('QE(electron) Factor ');
    title('Electrons available for photoemission by DOS vs E');
end

%Quantum Efficiency
QE = E_pe .* Fee .* (1-R2);

if(plot_all)
    figure(7)
    semilogy(lambda,QE);
    xlabel('lambda [nm] ');
    ylabel('QE(total) Factor ');
    title('Total QE vs lambda');
end

E_Excess = E-work_function;
if(plot_all)
    figure(8)
    semilogy(E_Excess,QE,'bo-');
    xlabel('E- workfunction [eV] ');
    ylabel('QE(total) Factor ');
    title('Total QE vs Excess Energy');
    set(gca,'Xlim',[0,1]);
end

%Normalized Emittance/Beam size [um*mrاد/nm]
enorm = 1e3*sqrt((E-phi_eff)./(3*0.511e6));
if(plot_all)
    figure(9)
    plot(E_Excess,enorm,'bo-');
    xlabel('E- workfunction [eV] ');
    ylabel('\frac{\epsilon_n}{\sigma_x} ');
    title('Emittance vs Excess Energy');
    set(gca,'Xlim',[0,1]);
end

end

```

A.3 Calculation of the Transmission Coefficient (Fresnel_T_coeff.m)

```

function [T] = Fresnel_T_Coeff(n,k,psi)
%This function calculates the Photoemission yield as a function of angle as reported
%in J. Hager, et al. Surface Science 600(2006) 1129-1133
%vs A. Gerlach, et al. Surface Science 492(2001) 214-224

```

```

%Gerlach uses the complex dielectric function as a fit parameter
%using a modified surface dielectric function to better fit the data on
%Cu(111)
%e_bulk(cu(111)) = 0.63+i*0.74
%e_surface(cu(111)) = 1.03 + i*0.22

n_complex = n+li*k;

epsilon = (n_complex).^2;

%Material Transmission Coefficient
t_real = sqrt(epsilon-(sin(psi).^2))./(epsilon.*cos(psi));
t_factor = (2*sqrt(epsilon).*cos(psi))./(epsilon.*cos(psi)+sqrt(epsilon-(sin(psi)).^2));
T = (abs(sqrt(epsilon)).^2).*real(t_real).*(abs(t_factor)).^2;
T_0 = T(psi==0);

if(0)
    figure(1);
    plot(psi.*180./pi,T./T_0,'r-');
    hold on;
    xlabel('Angle of Incidence [nm]');
    ylabel('T');
    title({'Transmission Coeff vs Angle','Hager'});
end

end

```

A.4 Junker-Waldron-Jaccodine AREY Theory (JWJ.m)

```

function [y] = JWJ(n,k,B,p,theta)
% This function calculates the photoelectric enhancement using JWJ theory
% n,k real and complex indices of refraction
% B - enhancement parameter
% p - degree of polarization (Iperp-Ipara)/I
% theta - range of angles
% "Vectorial Photoelectric Effect in Clean Molybdenum", J. Op. Soc Am
% 54,2(1964)

s0 = n + li*k;
s = sqrt((s0^2)-((sin(theta)).^2));

Cv = B*(n^2 + k^2)^2;

y0 = abs((s0+1)./(s+(s0^2).*cos(theta))).^2;
y1 = abs(s).^2;
y2 = abs((s+(s0.^2).*cos(theta))./(s+cos(theta))).^2;
y3 = Cv.*(sin(theta)).^2;

y = y0.*(cos(theta)/2).*((1+p)*y1 + (1-p)*y2 + (1+p)*y3);

end

```


A.5 AREY Fit Routine (fitJWJ.m)

```
function [estimates,model] = fitJWJ(xdata,ydata,n,k,p)
%This function fits the JWJ curve to a VPE Dataset
B_start = 1;
x0 = 1;
paramlist(1) = B_start;
paramlist(2) = x0;

model = @JWJfun;
estimates = fminsearch(model,paramlist);

function [sse,fitcurve] = JWJfun(params)
    B = params(1);
    x = params(2);
    fitcurve = JWJ(n,k,B,p,xdata);
    yerror = fitcurve - ydata;
    xerror = xdata(fitcurve==min(fitcurve)) - x;

    sse = sum(yerror)^2 + xerror(1)^2;

    if(0)
        figure(3)
        plot(xdata-x,fitcurve);
        hold all
    end

end

end
```

A.6 Time-of-Flight Analysis Codes (TOF_Analysis.m)

```
close all
clear all

accum_numevents = 0;

x_raw = zeros(0);
y_raw = zeros(0);
sum_x_raw = zeros(0);
sum_y_raw = zeros(0);

x_mm = zeros(0);
y_mm = zeros(0);
TOF_ns = zeros(0);

x_pixel_num=3044;
y_pixel_num=2902;
MCP_length=42e-3; %in m
x_pixel_m_factor=MCP_length/x_pixel_num ;
y_pixel_m_factor=MCP_length/y_pixel_num;
pixel_time=27e-12; %27ps
```

```

d = 36;%mm

t0 = 233;

xcenter = 28;
ycenter = 28;
for i=1:20
filepath = sprintf( '//Users//greaves//Documents//Research//Photoemission//←
Photoemission//Data//TOF_data//10232008//ONSAMPLE/GRID_GND//0.0V//10←
_23_test_214nm%d.bin ',i-1);

[data,numevents] = read_tof_file(filepath);
accum_numevents = numevents+accum_numevents;

[X_RAW,Y_RAW,SUM_X,SUM_Y] = read_tof_memory(data,numevents);
x_raw=double(vertcat(x_raw,X_RAW));
y_raw=double(vertcat(y_raw,Y_RAW));
sum_x_raw=double(vertcat(sum_x_raw,SUM_X));
sum_y_raw=double(vertcat(sum_y_raw,SUM_Y));

end

x_mm=x_pixel_m_factor*1e+3*x_raw-xcenter; %mm
y_mm=y_pixel_m_factor*1e+3*y_raw-ycenter; %mm
TOF_ns=pixel_time*1e+9*(sum_x_raw+sum_y_raw)./4 ; %ns
mintof=min(TOF_ns)
TOF_ns=TOF_ns - t0;

if(1)
%cuts
tmax = (1/3.8e6)*1e9;

mask = (sqrt((x_mm).^2 + (y_mm).^2)< 42) & (TOF_ns < tmax) & (TOF_ns > 0);
x_mm = x_mm(mask);
y_mm = y_mm(mask);
TOF_ns = (TOF_ns(mask)+45.5);
end

figure(1)
hold on
accum_numevents
plot(x_mm,y_mm,'r.')
xlabel('x(mm)')
ylabel('y(mm)')

figure(2)
hold on
hist(TOF_ns,20000)
xlabel('TOF(ns)')
ylabel('intensity(a.u.)')
min(TOF_ns)
max(TOF_ns)

if(1)
Ex = 1e6.*(1/2).*(0.511/(3e8)^2).*((x_mm.*1e-3)./(TOF_ns.*1e-9)).^2;
Ey = 1e6.*(1/2).*(0.511/(3e8)^2).*((y_mm.*1e-3)./(TOF_ns.*1e-9)).^2;

```

```

Ez = 1e6.*(1/2).*(0.511/(3e8)^2).*((d*1e-3)./(T0F_ns.*1e-9)).^2;
E = sqrt(Ex.^2 + Ey.^2 + Ez.^2);
Ebins = min(E):(mean(E)-min(E))/10000:3*mean(E);
figure(3);
Ehist=histc(E,Ebins);
bar(Ebins,Ehist,'histc');
xlabel('Energy(eV)');
ylabel('intensity (a.u.)');

kx = 1e-10*(0.511e6/(((3e8)^2)*6.57e-16)).*(x_mm.*1e-3)./(T0F_ns.*1e-9); %Angstrom
ky = 1e-10*(0.511e6/(((3e8)^2)*6.57e-16)).*(y_mm.*1e-3)./(T0F_ns.*1e-9); %Angstrom

kxbins = (-0.2:0.4/10000:0.2);
figure(4);
kxhist=histc(kx,kxbins);
bar(kxbins,kxhist,'histc');
xlabel('kx(1/angstrom)');
ylabel('intensity (a.u.)');

kybins = (-0.2:0.4/10000:0.2);
figure(5);
kyhist=histc(ky,kybins);
bar(kybins,kyhist,'histc');
xlabel('ky(1/angstrom)');
ylabel('intensity (a.u.)');

%E vs kx
mask = (ky < 0.01)&(ky>-0.01);
kxmask = kx(mask);
Emask = E(mask);

Exhist = figure(6);
axhist = gca;
xy2Dhist(Exhist,axhist, kxmask, Emask, -.25:.001:.25,0:0.01:1,{ 'E vs k_x', 'k_x [1/↵
Angstrom] ', 'E [eV] '}, 'plot');

%E vs ky
mask = (kx < 0.01)&(kx>-0.01);
kymask = ky(mask);
Emask = E(mask);

Eyhist = figure(7);
ayhist = gca;
xy2Dhist(Eyhist,ayhist, kymask, Emask, -.25:.001:.25,0:0.01:1,{ 'E vs k_y', 'k_y [1/↵
Angstrom] ', 'E [eV] '}, 'plot');

%kx vs ky
Ecenter = 0.8;
Ewidth = 0.02;
mask = (E < Ecenter + Ewidth)&(E>Ecenter - Ewidth);
kymask = ky(mask);
kxmask = kx(mask);

Eyhist = figure(8);
ayhist = gca;
xy2Dhist(Eyhist,ayhist, kxmask, kymask, -.25:.001:.25,-.25:.001:.25,{ 'k_y vs k_x',↵
'k_x [1/Angstrom] ', 'k_y [1/Angstrom] '}, 'plot');

end

```

```
if(0)
    for i = 1:8
        filepath = sprintf('//Users//greaves//research//outfiles//figure%d.eps',i)↵
        ;
        print(i, '-depsc',filepath);
    end
end
```

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