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Spectral Response of Rejuvenated Cs-Te Photocathodes for Optimized Production Control

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Master's Thesis

Master's Degree Programme: Chemical Engineering

Department of Chemical Process Engineering
Mannheim University of Applied Sciences

30.11.2024

Conducted at:

European Organization for Nuclear Research (CERN),
Geneva, Switzerland

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Declaration

With this statement I, Palma Katona, declare that I have independently completed this master's thesis titled "Spectral Response of Rejuvenated Cs-Te Photocathodes for Optimized Production Control" and have not used any other sources other than those indicated in the thesis. All thoughts and illustrations that are taken directly or indirectly from external sources are properly marked as such. The thesis has not been submitted in this or a similar form to any other academic institution.

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Abstract

Precise control of the Cs-Te stoichiometric deposition ratio during the photocathode fabrication process is essential to optimize the resulting quantum efficiency (QE) and emittance of electron sources. For example, an excess of Cs or the formation of another Cs - Te phase can lead to a sub-optimal electron bunch energy spread or a decrease in overall quantum yield. This study investigates the spectral response characteristics of Cs - Te photocathodes rejuvenated by co-depositing a Cs - Te layer over a degraded one. For this, the QE of the photocathodes was measured using a 10 Hz 5 ns pulsed OPO in the wavelength range from 240 to 430 nm. This experiment allows comparison of the spectral response of rejuvenated photocathodes with non-rejuvenated ones reported in the literature, facilitates the estimation of the photoemission threshold for the photoemissive phase present on the cathode, and allows comparison with the expected spectral response predicted by Spicer's Three-Step Model. Our results contribute to a deeper understanding of the fabrication of Cs-Te photocathodes and provide practical insights for optimizing production to improve performance.

Acknowledgements

I would like to express my sincere gratitude to my supervisor, Ralf Erik Rossel, for his guidance, support and encouragement. His expertise and advice were greatly valued throughout this project.

I would also like to thank Dr. Anabel Clemen, my university supervisor, for supervising this thesis and providing the necessary academic support throughout the process.

I am especially grateful to Eduardo Granados for his assistance and sharing of his knowledge during the course of this thesis. His insights and willingness to help were essential to the success of this research.

I would like to thank Cyril Bernerd and Miguel M. Calderon for their assistance during the experimental phase. Their technical expertise contributed greatly to my understanding of the experimental setup and measurement techniques.

I would also like to thank the CRIS collaboration team for providing access to their OPO, which was crucial for the measurements performed in this thesis.

I would like to thank the entire LP group for their feedback and support throughout this project. It has been a great experience working with such a knowledgeable and collaborative team.

Finally, thank you to my friends and family for their encouragement and support throughout this journey, and Yehia for his constant support every step of the way.

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Abbreviations

AWAKE – Advanced Wakefield Experiment

CLEAR - CERN Linear Electron Accelerator for Research

DC – Direct Current

FC – Faraday Cup

FCT – Fast Current Transformer

FEL – Free-Electron Laser

LINAC – Linear Accelerator

MTE – Mean Transverse Energy

NEA – Negative Electron Affinity

ND – Neutral Density

OPO – Optical Parametric Oscillator

PEA – Positive Electron Affinity

PVD – Physical Vapor Deposition

QE – Quantum Efficiency

RF – Radio Frequency

SH – Second Harmonic

SF – Sum Frequency

TESS – Transverse Energy Spread Spectrometer

TSM – Three Step Model

UHV – Ultra-High Vacuum

WCM – Wall Current Monitor

XPS – X-ray Photoelectron Spectroscopy

XRD – X-ray Diffraction

CYL – Cylindrical Current Measurement Device

Chapter 1

Introduction

This chapter provides an overview of photocathodes and their role in electron production for particle accelerators. It discusses the principles of photocathode technology, its advantages over other electron sources, and its applications.

1.1 The CERN Accelerator Complex

CERN, the European Organization for Nuclear Research—located in Meyrin, Switzerland—is one of the world’s largest nuclear and particle physics research organisations. Since its foundation in 1954, CERN has pushed the boundaries of physics and made discoveries that have reshaped our understanding of the universe [1, 2]. At its heart lies the world’s largest and most powerful particle accelerator, the Large Hadron Collider (LHC). The primary purpose of the LHC was to confirm the existence of the Higgs boson, a fundamental particle responsible for giving mass to particles, as predicted by the Standard Model. This milestone was achieved in 2012, marking one of the most significant discoveries in modern physics. [3] The LHC is a 27-kilometer-long accelerator located beneath the French-Swiss border and is used to collide particles at high energies, simulating conditions similar to those after the Big Bang [2, 4].

At the LHC, particles such as protons or heavy ions are accelerated to almost the speed of light after being injected into its two counter-rotating beamlines, where they are guided along their trajectories by powerful superconducting magnets [5]. The acceleration process takes place in several stages as the particles pass through successive

loops of accelerators to achieve higher velocities. This process begins with the linear accelerators (LINACs), followed by a series of circular accelerators of increasing size in each loop—the booster, PS booster, SPS, and finally the LHC itself [6].

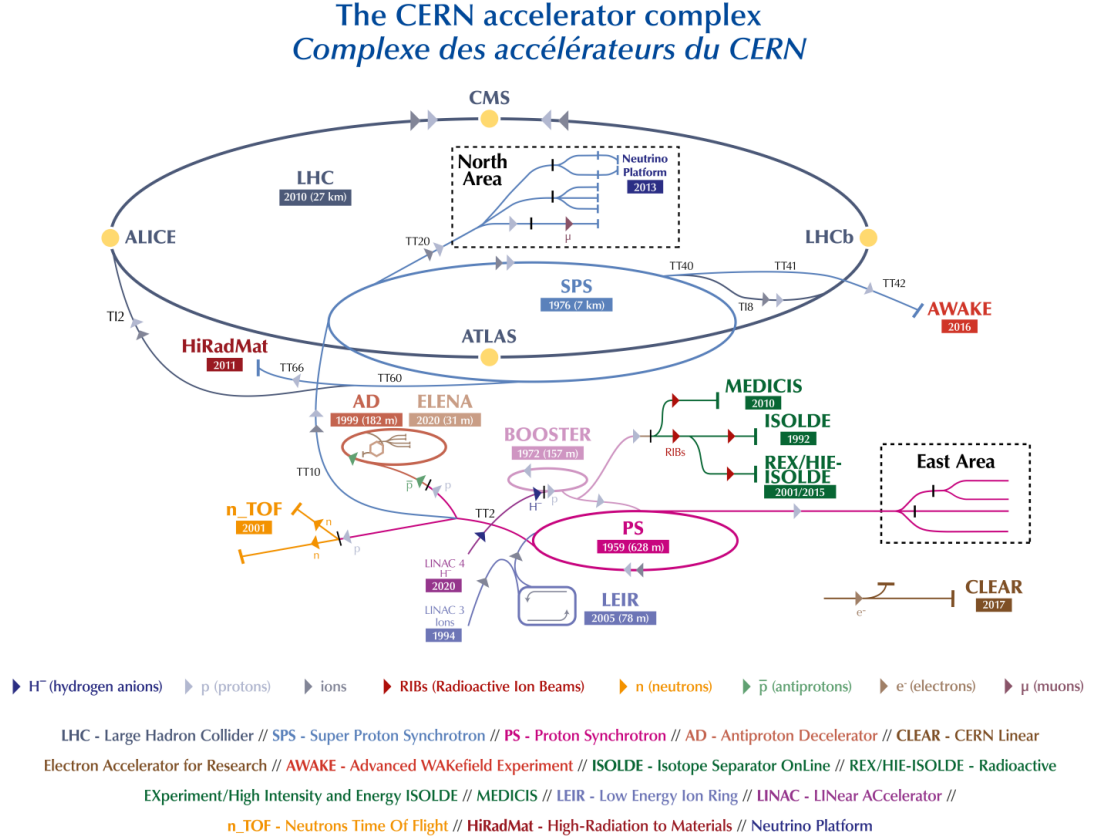


FIGURE 1.1: CERN accelerator complex [6].

The accelerated particles are then directed to collide at four interaction points along the accelerator ring: ATLAS, CMS, LHCb, and ALICE. These collision points are instrumented with sophisticated detectors that allow scientists to observe and analyze the resulting particle interactions in detail [5].

CERN has been a pioneer in many technological innovations, including advances in computing, data analysis techniques, and detector technologies. These innovations have not only supported the operation of the LHC but have also found applications in fields such as medical imaging, materials science, and environmental monitoring and protection [7].

From discovery machines like the LHC, CERN is now making a strategic transition to precision machines like the Compact Linear Collider (CLIC) and the Future Circular Collider (FCC). Unlike the LHC, which is designed to discover new particles and phenomena, these precision machines focus on making highly accurate measurements of known particles. This is crucial for testing the Standard Model and detecting subtle deviations that may point to new physics. For example, in its initial phase, the FCC-ee will study electron-positron collisions at very high energies, addressing questions about dark matter and the matter-antimatter asymmetry in the universe [8]. Achieving the level of precision required in these machines depends heavily on the quality of the electron beams. In electron-positron colliders such as the FCC-ee, high luminosity is essential for precision measurements. This in turn requires electron beams with high bunch charges and good beam stability. The FCC-ee design specifies a bunch charge of about 6.7 nanocoulombs (nC), corresponding to 4.2×10^{10} electrons per bunch [9].

1.2 Electron Sources

The growing emphasis on electron-based science, as demonstrated by CERN's proposal to build the FCC, is driving significant advancements in electron source technologies. Such experiments require electron sources capable of producing high-quality beams with low emittance and high bunch charges. Different electron generation methods produce electron bunches with varying parameters. The three primary electron gun technologies are based on thermionic emission, field emission, or photoemission. Thermionic guns, produce electron beams by applying thermal energy to the material, allowing electrons to overcome the material's energy barrier (work function Φ) and escape into vacuum. Field emission sources, on the other hand, rely on the quantum mechanical tunneling of electrons through the potential barrier of a material when a strong electric field is applied. In contrast, photoinjectors use photocathodes to generate electron beams through the process of photoelectric effect. In this process, photons from a light source, typically a laser, supply the energy needed for electrons to overcome the material's threshold energy and be emitted into the vacuum [10, 11]. Their emission models can be seen in Figure 1.2.

Selecting the electron source for a particular application depends on several criteria related to the properties of the electron beam, including thermal emittance, beam brightness, current density and pulse length [10]. Emittance refers to the spread of

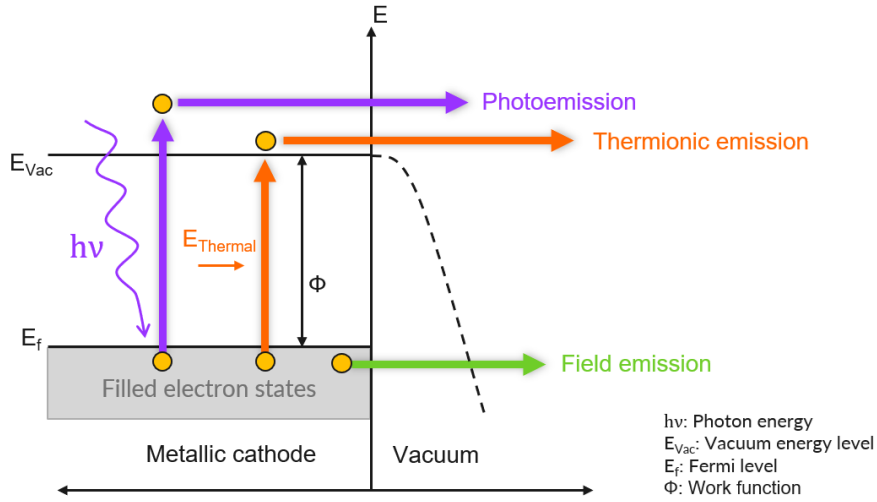


FIGURE 1.2: Energy-band diagram of thermionic emission, field emission and photoemission mechanisms (Figure modified from [12]).

the electron beam in phase space, combining its position and momentum distributions. A lower emittance indicates a more confined beam, resulting in more precise electron bunches. This is critical for applications such as free-electron lasers (FELs), where low emittance improves the coherence and intensity of the generated photons [13]. Current density measures the amount of current emitted per unit area of the source. High current densities are essential for applications such as medical accelerators, where electron beams must generate intense X-rays for cancer radiotherapy [14]. Pulse length determines the ability to produce short, well-defined electron bunches, which is critical for time-resolved experiments such as ultrafast electron diffraction, where it is necessary to capture dynamic processes on femtosecond timescales [15]. Finally, beam brightness combines two parameters, beam current and emittance. High brightness is critical for particle accelerators such as the Future Circular Collider, where precision electron-positron collisions depend on intense and low-emitting beams [16].

Since 1985, photoinjectors have gained prominence due to their significant advantages. Photocathodes are now the preferred electron source in many modern accelerators due to their superior performance in terms of emittance and brightness compared to thermionic guns. They can deliver high current densities, exceeding 200 A/cm², without being destroyed, and their electron beams can be precisely modulated by the driving laser pulses. This enables the generation of short, well-defined electron bunches. Another major advantage is the lower beam emittance achievable with photoinjectors.

Historically, thermionic guns struggled to achieve beam emittances below 20 microns for a 1 nC bunched beam. In contrast, the first photocathode RF guns, introduced in the late 1980s, achieved emittances around 10 microns, and by 2010, emittances as low as 1 micron had been demonstrated, marking a significant improvement in beam quality for advanced applications (see Figure 1.3) [10, 17].

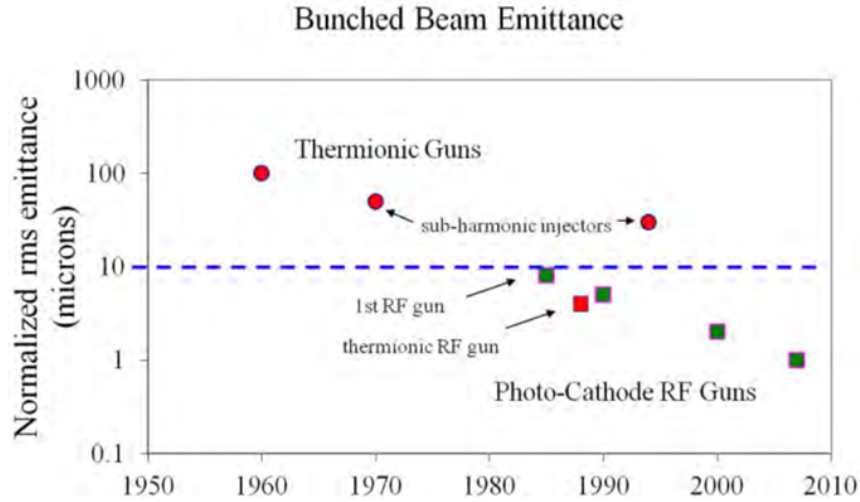


FIGURE 1.3: History of the 1 nC bunched beam emittance over the past 60 years [10].

A photoinjector consists of a laser system, a photocathode, and an RF or a DC gun that generates the electric field required for electron acceleration. The basic structure of a photoinjector is illustrated in Figure 1.4.

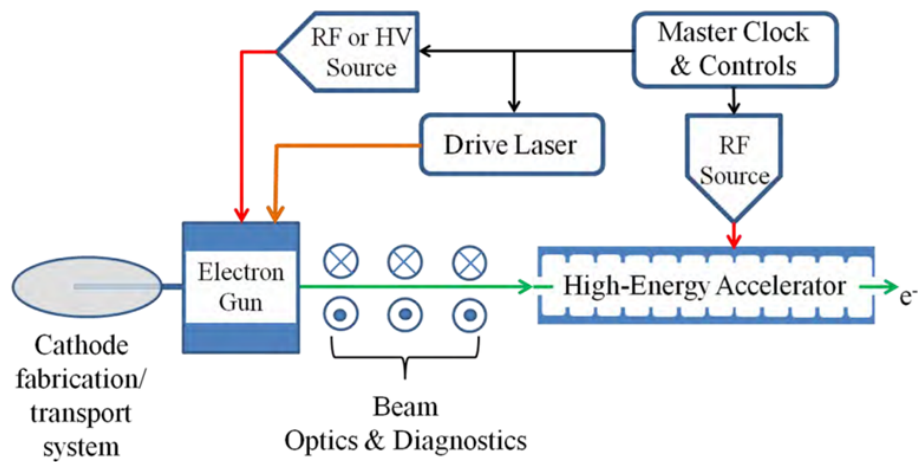


FIGURE 1.4: Fundamental components and structure of a photoinjector system [10].

Photoinjectors play a central role in electron accelerators, not only generating the electron beam, but also precisely delivering it to the linear accelerator (LINAC) for further acceleration. The process begins with the photocathode being illuminated by a pulsed laser, triggering electron emission via the photoelectric effect. The emitted electrons are then accelerated by the electron gun, which can be powered by either a conventional or superconducting RF system or a high-voltage DC source, depending on the design. These systems create the necessary field gradient to accelerate the electrons along the beam path. After acceleration, the photoinjector's optical system focuses and directs the electron beam, ensuring that it has the proper size and shape for efficient injection into the high-energy accelerator. The electric field generated by the RF gun also significantly influences the beam emittance and bunch duration, which are critical parameters for accelerator performance. The overall performance of a photoinjector depends on the characteristics of each of its components [10].

1.3 Photocathodes at CERN

Photocathodes are devices used for generating electron beams in accelerator applications where short bunches and high peak intensities are required. At CERN, they are used in two linear accelerator experiments: the CERN Linear Electron Accelerator for Research (CLEAR) and the Advanced Wakefield Experiment (AWAKE).

The production and R&D of photocathodes for these experiments are carried out in CERN's Photoemission Laboratory, which was founded in 1989 with the purpose of serving as a test facility for CERN's Compact Linear Collider (CLIC) experiment [18, 19]. Although CLIC, an electron-positron accelerator designed for high-energy studies of the Higgs boson, has not yet been built [20], the laboratory has remained operational since its inception. Over the years, it has focused on extensive research and development in the field of photoinjectors and the advancement of photocathodes. In addition to its R&D work, the laboratory has also taken over the production of photocathodes for subsequent programs such as the CLEAR and the AWAKE experiments in order to ensure a continuous supply and maintenance of photocathodes for these facilities [21, 22].

CLEAR is a user facility designed to develop components for current and future accelerators, while exploring innovative concepts such as THz and plasma acceleration.

In addition, CLEAR provides beams for medical applications, such as FLASH radiotherapy for cancer treatment, and is conducting studies on the radiation hardness of electronic components, important for aerospace applications [23].

AWAKE, the Advanced Wakefield Experiment, is the world's first proton-driven plasma wakefield accelerator. Protons from CERN's SPS ring are injected into a 10 m long plasma cell, creating a wakefield. An electron beam is then injected into this wakefield, accelerating the electrons to energies up to several gigavolts. AWAKE aims to develop compact, high-energy accelerators by achieving high energy gains over short distances [24, 25].

1.4 Applications of Photoinjectors

Electron accelerators using laser-driven photoelectron emission have a wide variety of applications. One of the key uses is as an electron source in advanced research technologies such as free-electron lasers (FELs) and X-ray diffraction [26]. FELs can produce coherent radiation at wavelength ranges that are otherwise difficult to achieve, particularly in the sub-10 nm range [27]. This opens up new possibilities for studying atomic and crystalline structures in areas such as X-ray diffraction, where short wavelengths of X-rays are used to probe the arrangement of atoms in materials. The X-rays are reflected by the atoms in the material, producing a diffraction pattern that provides valuable information about the atomic structure. The technique is non-destructive and can be applied to a wide range of materials, including metals, polymers, ceramics, and semiconductors [28].

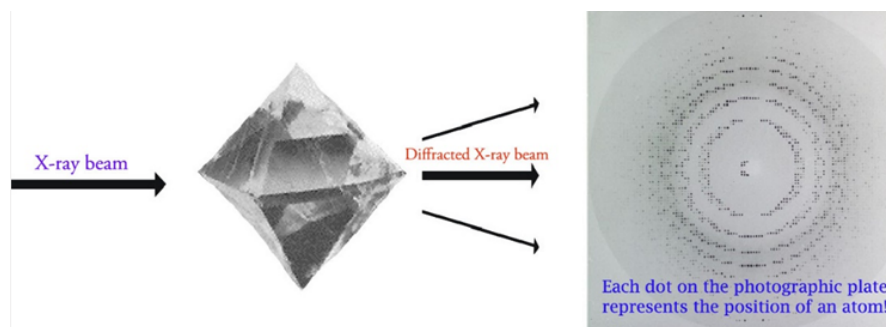


FIGURE 1.5: X-ray diffraction pattern of a crystal [29].

Electron accelerators have also emerged as a promising tool for medical treatment, particularly in the field of oncology. The process of irradiating tumor cells with an

electron beam is called FLASH therapy, in which a very high dose of radiation is delivered to the patient in a few nanoseconds. It is proven to be very effective in targeting tumor cells while minimizing damage to surrounding tissues [30, 31]. These applications highlight the importance of producing high quality electron beams that rely on advanced photoinjectors to optimize beam quality in terms of brightness, emittance, bunch charge, and pulse duration.

Chapter 2

Theory of Photoemission

This chapter provides an overview of the photoelectric effect, beginning with its historical context in Hertz's experimental discoveries and Einstein's theoretical formulation. The chapter then introduces the quantum model of photoemission and explains how this framework improves our understanding of the phenomenon. To evaluate photoemission yield, the quantitative measure known as quantum efficiency is presented. Furthermore, we will describe photoemission using Spicer's Three-Step Model, interpreting it in terms of the band theory of metals and semiconductors.

2.1 The Photoelectric Effect

In 1887, Heinrich Hertz announced the discovery of the photoelectric effect. Hertz's experiment was originally designed to provide experimental confirmation of Maxwell's theoretical predictions about electromagnetic waves. His apparatus consisted of an electric circuit to produce a spark and another circuit to receive it. In between is a spark gap. When a current is generated in the first circuit, the spark should travel through the gap and be transmitted to the second circuit, where it could be observed in the spark gap of the second circuit, indicating the presence of electromagnetic waves. To detect the spark in the second circuit more clearly, he shielded the apparatus with a dark box. He found that producing a spark in the secondary circuit became more difficult in the absence of light. Further investigation revealed that ultraviolet (UV) light emitted by the initial spark played a crucial role in facilitating electron emission in the secondary

circuit. Hertz found that the dark box absorbed this UV light, preventing the effect. By testing different shielding materials, including glass, horn, and crystals like rock salt, Hertz confirmed that UV radiation from the primary spark was responsible for the phenomenon. His observations demonstrated that ultraviolet photons emitted by the primary spark caused electrons to be ejected from the metal plate in the secondary circuit, a phenomenon now known as the photoelectric effect [32]. This observation challenged the classical wave theory of light and provided the basis for the development of quantum mechanics. In 1905, Albert Einstein extended Hertz's findings by proposing that light consists of discrete packets of energy called photons. This explanation of the photoelectric effect provided direct evidence for the quantisation of light and transformed our understanding of atomic and subatomic processes. Einstein's work on the photoelectric effect earned him the Nobel Prize in Physics in 1921 [33]. The quantum model of photoemission is shown in Figure 2.1.

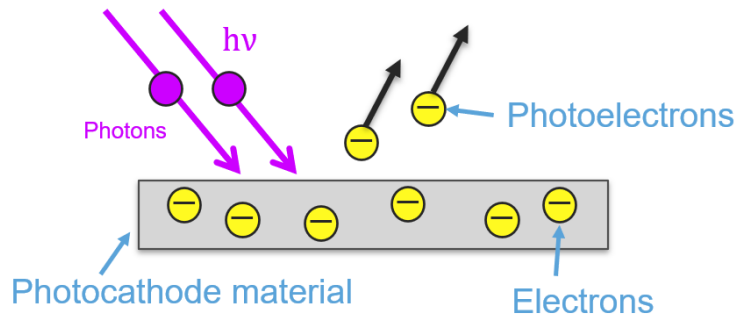


FIGURE 2.1: Schematic representation of the photoelectric effect, illustrating the ejection of an electron from the surface of a material when exposed to incident photons with an energy of $(h\nu)$.

In the quantum model, a photon transfers its energy to an electron, enabling it to overcome the material's energy barrier and escape into the vacuum. For the electron to be emitted, the energy of the incoming photon must be greater than or equal to the threshold energy of the material, which is the minimum energy required to release the electron from the surface. If the photon energy is sufficient, the electron will be ejected from the material [34]. The energy of a photon E can be expressed as:

$$E = \hbar\omega = h\nu = \frac{hc}{\lambda}, \quad (2.1)$$

where h represents Planck's constant (6.626×10^{-34} J·s), and $\hbar$ represents the reduced Planck's constant, defined as $\hbar = \frac{h}{2\pi}$. In this context, ω represents the angular frequency of the photon, where $\omega = 2\pi\nu$, and ν is the frequency of the photon (in Hz). c refers to the speed of light in a vacuum (3.00×10^8 m/s), and λ is the wavelength of the photon (in nm) in vacuum. The number of electrons emitted is proportional to the number of photons impinging on the surface. As the intensity of the light increases (i.e., the number of incident photons increases), more electrons can be ejected, resulting in a higher total emitted current [34].

A quantitative measure of photoemission is the quantum efficiency (QE), defined as the ratio of the number of electrons emitted to the number of photons impinging on the surface. This measure is described by the Fowler-Dubridge equation and can be expressed using Planck's constant h , the speed of light c , the elementary charge q_e , the incident laser wavelength λ , the total emitted charge ΔQ , and the total energy of photons impinging on the surface ΔE [35, 36]

$$QE = \frac{\text{Number of electrons}}{\text{Number of photons}} = \frac{h\nu \Delta Q}{q_e \Delta E} . \quad (2.2)$$

Summarizing all the constants in Equation 2.2, the quantum efficiency can be expressed in a simplified form as follows [17]:

$$QE = 124 \frac{\Delta Q [\text{nC}]}{\lambda [\text{nm}] \Delta E [\mu\text{J}]} . \quad (2.3)$$

This calculation assumes that the illumination time is significantly longer than the time required for photon absorption and electron transport within the material, allowing the simplification that electron emission occurs almost simultaneously with surface illumination. It also assumes that the illumination intensity is not high enough to cause significant heating of the metal, ensuring that photoemission is not affected by thermal emission [26, 35].

2.2 Band Theory of Materials

To understand the mechanisms behind photoemission, we first need to study the band structure of metals and semiconductors. An overview of the band structure of materials is presented in Figure 2.2.

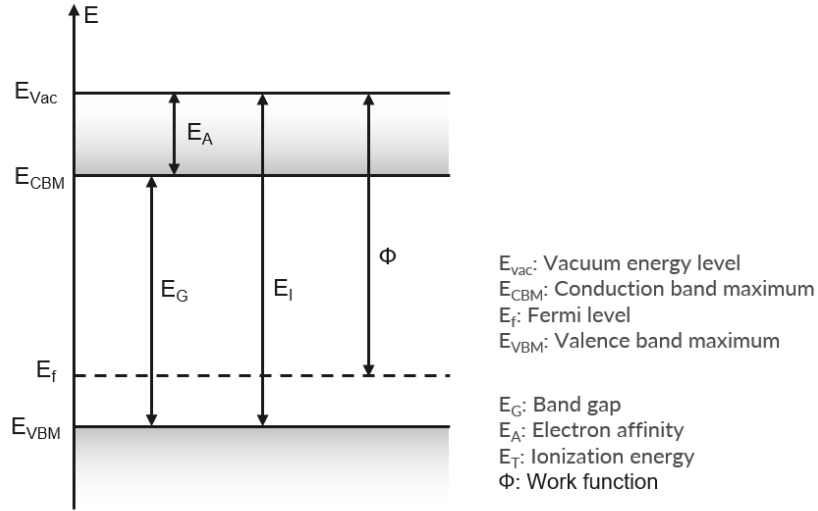


FIGURE 2.2: Schematic representation of the band structure of materials (Figure modified from [36]).

The highest energy state that an electron can occupy within the valence band is defines as the valence band maximum (E_{VBM}). The lowest unoccupied energy level in the conduction band corresponds to the conduction band minimum (E_{CBM}). The energy gap between these two levels is known as the band gap (E_G), a region where electron states are forbidden. Between the conduction band and the valence band is the Fermi level (E_F), which represents the highest occupied electronic state at absolute zero. At this temperature, the Fermi level marks the boundary between the occupied and unoccupied electronic states, although it is not necessarily within the band gap for metals or heavily doped semiconductors. In a thermodynamic context, the Fermi energy is often referred to as the chemical potential (μ). The vacuum energy level (E_{Vac}) defines the potential energy required for an electron to escape from the solid. It represents the energy of an electron positioned just outside the metal's surface, where its kinetic energy is zero. The difference between the conduction band minimum and the vacuum energy level is called the electron affinity (E_A), while the difference between the valence band maximum and

the vacuum level is called the ionization energy (E_I). The ionization energy represents the minimum energy required to remove an electron from the valence band to the vacuum energy level. The photoemission threshold energy (E_T) is closely related to the ionization energy, as both describe the energy required to remove an electron from a material. In metals, the threshold energy corresponds to the work function (Φ), which is the energy required to remove an electron from the Fermi level (E_F) to the vacuum level (E_{Vac}). In semiconductors, the threshold energy is determined by the sum of the bandgap energy (E_G) and the electron affinity (E_A), which accounts for the energy required to excite an electron from the valence band to the conduction band and then release it into the vacuum [36, 37]

$$E_T = E_G + E_A . \quad (2.4)$$

In metals, photoelectrons originate from the Fermi level and must overcome the material's work function, which is a fixed value. In semiconductors, the work function can vary as the Fermi level depends on the type and degree of doping. The electron affinity remains independent of doping [38].

The electrical conductivity of metals and semiconductors exhibits significant differences due to variations in their band structures. Semiconductors are characterized by the presence of a band gap, whereas metals have no band gap because the valence band overlaps with the conduction band, allowing free electrons to move to the conduction band. The band gap in semiconductors typically ranges from 1 to 4 eV and represents the energy required for an electron to be promoted from the valence band to the conduction band, allowing electrical conduction in the material. At absolute zero, all states below the Fermi energy are occupied, while those above are empty. In semiconductors, as the temperature rises above 0 K, some electrons gain enough energy to overcome the band gap and move from the valence band to the conduction band. As a result, the conduction band becomes partially filled, allowing these electrons to contribute to electrical conductivity. Metals behave differently as their Fermi level is within the conduction band. This means that even at low temperatures, a significant number of free electrons are available for conduction. This makes metals inherently good conductors. The difference between the band structure of semiconductors, and metals is shown in Figure 2.3 [37, 39].

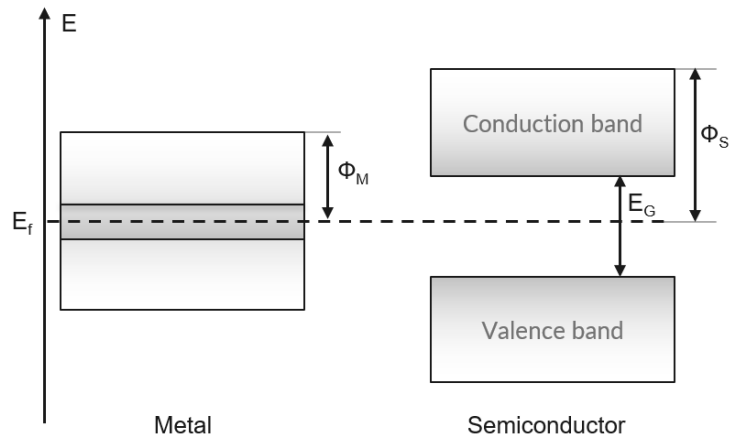


FIGURE 2.3: Energy band diagram of metals and semiconductors, , where Φ_M is the work function of metals, Φ_S is the work function of semiconductors, E_G is the semiconductor band gap and E_F is the Fermi energy (Figure modified from [40]).

The Fermi level in semiconductors varies with the type and degree of doping, as doping directly changes the charge carrier concentration. In an undoped, or intrinsic, semiconductor, there are no impurities, so the electrons and holes are in equilibrium. Each electron that is promoted to the conduction band leaves a corresponding hole in the valence band. At absolute zero (0 K), the Fermi level of an intrinsic semiconductor is exactly in the center of the band gap, but it can shift with increasing temperature as electrons gain energy to move across the band gap. In contrast, extrinsic semiconductors contain intentional impurities, or dopants, to increase the concentration of either electrons or holes. N-type semiconductors are created by adding donor atoms that introduce extra electrons. This excess of electrons moves the Fermi level closer to the conduction band, reflecting the greater likelihood of electrons occupying energy states in the conduction band. On the other hand, p-type semiconductors are created by adding acceptor atoms, which introduce positive charge carriers into the material, which then create holes. The Fermi level shifts closer to the valence band due to the altered charge distribution and results in a higher probability of finding electrons in the valence band. An element can be either a donor or an acceptor, depending on the lattice in which it is embedded [37]. A visual representation of the different charge distributions in intrinsic, n-type, and p-type semiconductors is shown in Figure 2.4.

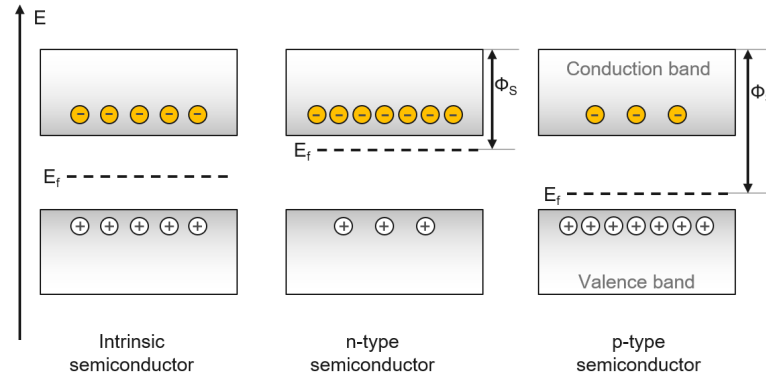


FIGURE 2.4: Band diagram of intrinsic, n-type, and p-type semiconductors (Figure modified from [40, 41]).

The band gap of semiconductors can be determined by studying how they absorb light at different energies. If the energy of the incoming photon is greater than the band gap, it excites electrons from the valence band to the conduction band, resulting in the absorption of light. However, if the photon energy is less than the band gap, there isn't enough energy to excite the electrons and the light passes through the material. By observing the energy at which light is absorbed, the band gap can be accurately measured [37].

2.3 The Three-Step Model for Photoemission

Current photoemission models provide a comprehensive picture for understanding the underlying physical mechanisms of photoemission. Spicer's Three-Step Model (TSM) breaks down photoemission into three distinct steps. This allows a more detailed analysis of the phenomenon at the atomic level [36]. The three steps are illustrated in Figure 2.5.

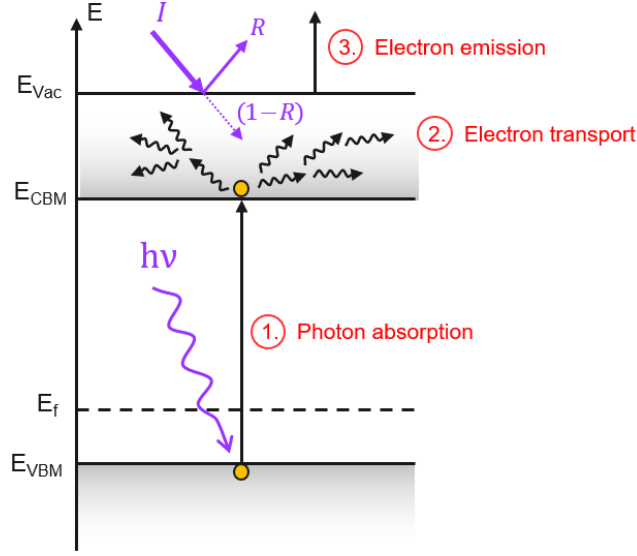


FIGURE 2.5: Schematic Representation of Spicer's Three-Step Model of Photoemission, where I represents the incident light intensity, R the reflected portion, and $1 - R$ the transmitted or absorbed portion. $h\nu$ denotes the energy of a single photon. (Figure modified from [36]).

The first step is photon absorption. Photoexcitation drives electrons from the valence band into the conduction band, creating electron-hole pairs. For photoemission to occur, the photons must have sufficient energy to excite the electrons. The second step involves electron transport, where the photoexcited electrons migrate from the bulk of the material to its surface. During this process, they may experience scattering events such as electron-electron and electron-phonon interactions. The final step is electron emission, where the electrons that reach the surface must overcome the surface barrier to escape into the vacuum. The quantum efficiency in this model is determined by the product of the probabilities of these three steps [26, 35].

For metals, the QE, according to the three-step model, can be expressed with the following equation [35]:

$$QE = \frac{(1 - R)}{(1 + p)} \cdot \frac{\mu(\hbar\omega - \phi)^2}{4\hbar\omega(\mu + \hbar\omega)(2\mu - \hbar\omega)}, \quad (2.5)$$

where $(1 - R)$ represents the fraction of incident light that is absorbed (with R being the reflectivity), and p denotes the emission probability.

The emission probability can be described as

$$p(E) = \frac{\delta(\omega)}{l(E)} = \frac{m\delta(\omega)}{\hbar k(E)\tau(E)} , \quad (2.6)$$

where δ is the laser penetration depth, and $l(E)$ is the electron mean free path, which is a function of the total electron energy. The mean free path $l(E)$ can be expressed as:

$$l(E) = \frac{\hbar k \tau}{m} , \quad (2.7)$$

where k is the electron wave vector, τ is the relaxation time, and m is the electron rest mass ($\approx 510999 \text{ eV}/c^2$).

The wave vector k can be further described as

$$k = \frac{\sqrt{2mE}}{\hbar} . \quad (2.8)$$

For semiconductors, the QE is calculated with the following equation:

$$QE(\hbar\omega) \approx \frac{B}{g + \frac{g}{(\hbar\omega - V_g)^v}} , \quad (2.9)$$

where V_g is the sum of the material's band gap and the electron affinity. Spicer found that the parameter v is equal to $3/2$. In this model, B , g , and V_g are treated as adjustable fitting parameters to match the experimental data [35].

The second step in the Three-Step model is determining the energy and number of electrons that can escape. Firstly, only electrons traveling in an angular cone shape can escape, with a maximum angle determined by the electron's energy relative to the barrier height [35, 42]. Furthermore, the scattering events during electron transport also have a significant impact on the energy and number of escaping electrons. These events cause electrons to lose part of their energy, resulting in a greater energy spread among the photoelectrons. If they lose too much energy, they will not be able to overcome the potential barrier at the surface, reducing the number of photoelectrons that successfully reach the vacuum level and escape. Consequently, the scattering processes affect the quantum efficiency by influencing both the number and the energy distribution of the emitted photoelectrons [26].

The effect of scattering events during the electron transport phase differs significantly between metals and semiconductors. Metals lack a well-defined band structure, which leads to frequent scattering as electrons travel toward the surface. These scattering events, including electron-electron and electron-phonon interactions, cause many electrons to lose energy either partially or completely. Often, electrons lose all their energy due to these numerous scattering events, and as a result, these electrons cannot escape into the vacuum, contributing to a lower quantum efficiency. The mean free path of electrons in metals is typically very short, usually limited to the immediate surface layer, so only electrons very close to the surface are able to escape [36].

In contrast, semiconductors have a well-defined band structure that allows electrons to be excited from the valence band to the conduction band with minimal scattering. Once in the conduction band, electron-electron scattering is rare, resulting in a longer mean free path for the photoelectrons and consequently a higher quantum efficiency. Within a specific range of photon energies, known as the "magic window," as described by Spicer and Herrera-Gomez [36], electron-electron scattering is particularly minimized. In this energy range, the excitation conditions allow electrons to move through the conduction band with minimal interactions and energy loss. Lattice (phonon) scattering still occurs but has no effect on the QE due to the relatively small energy losses involved [35].

Chapter 3

Photocathodes

This chapter provides an overview of photocathodes, discussing photocathode materials, the fabrication process and methods of photocathode rejuvenation. Additionally, the effects of impurities on photocathode performance are explored.

3.1 Photocathode Materials

There are two main categories of photocathodes: metallic and semiconductor. Metallic photocathodes are commonly used in low- to medium-current applications and are typically made from materials like copper or magnesium, with a polished surface. These materials are favored for their robustness and long operational lifetime, making them suitable for applications where durability is essential. Metallic cathodes generally have a high work function, typically in the range of 4-5 eV, which can help to minimize the dark current, but requires high energy input and therefore the use of a UV driving laser. A limitation of metallic cathodes is their low quantum efficiency (QE), typically less than 1% [10].

For high average current applications, alkali-semiconductor photocathodes are the preferred choice, offering high quantum efficiency in the UV to visible range compared to metallic cathodes. However, they are highly sensitive to impurities, residual gas poisoning and low vacuum conditions, which can significantly reduce their lifetime. The lifetime of a semiconductor cathode is typically defined as the time it takes for its QE to drop to $1/e$ of its initial value. Commonly used alkali-based compounds in

semiconductor photocathodes include materials with tellurium or antimony and they can be categorized based on their composition. There are single-alkali cathodes such as cesium telluride (Cs_2Te) and cesium antimonide (Cs_3Sb), as well as bi-alkali cathodes like sodium-potassium antimonide (Na_2KSb) and multi-alkali cathodes, for example, cesiated sodium-potassium antimonide ($\text{Na}_2\text{KSb:Cs}$). Each type of material exhibits distinct spectral sensitivities, as illustrated in Figure 3.1 [10]. Additional details about common semiconductor cathodes are provided in Table 3.1, which summarizes key parameters such as quantum efficiency, threshold energy, and thermal emittance. While the QE and emittance values listed may not apply to every cathode in all cases, they provide a reliable approximation of the typical range.

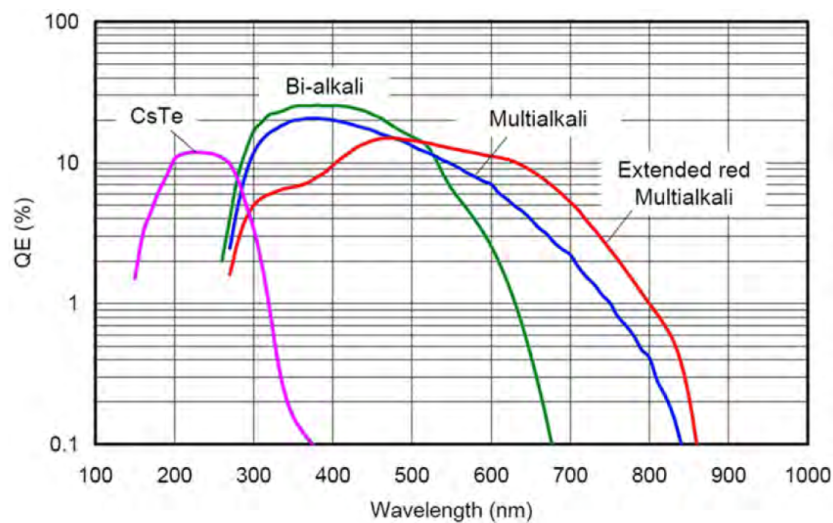


FIGURE 3.1: Typical spectral response of different alkali-photocathode materials [10].

As shown in Figure 3.1 and Table 3.1, antimony-based photocathodes typically offer good quantum efficiency with threshold energies in the range of 2-3 eV, allowing the use of a green laser as the driving source. Cs_2Te has a higher threshold energy of 3.5 eV, requiring the use of a UV laser for photoemission. While this requirement may make its implementation more challenging, Cs_2Te is widely favored due to its exceptional resilience to contamination by residual gases such as O_2 and CO_2 , compared to other semiconductor materials. This property ensures reliability for applications requiring long-term, high-performance operation. Furthermore, with a typical QE value of around 10%, as shown in Table 3.1, Cs_2Te provides sufficiently high efficiency for most practical applications [10].

TABLE 3.1: Parameters for commonly used semiconductor photocathodes, regarding their required photon energy for emission, quantum efficiency and characteristic thermal emission [10].

Cathode Wavelength	λ [nm] , E_{ph} [eV]	QE [%]	$E_a + E_{gap}$ [eV]	Thermal emittance $\left[\frac{\text{mm mrad}}{\text{mm rms}} \right]$	
				Theory (Equ. 7.6)	Experiment
Cs ₂ Te	262, 4.73	~10	3.5	0.9	1.2 ± 0.1
Cs ₃ Sb	532, 2.33 473, 2.62 405, 3.06	~4 ~7 ~9	1.6 + 0.45	0.42 0.62 0.82	0.56 ± 0.03 0.66 ± 0.03 0.80 ± 0.04
Na ₂ KSb	330, 3.76	~10	1 + 1	1.07	N/A
Na ₂ KSb:Cs	390, 3.18	~20	1 + 0.55	1.03	N/A
K ₂ CsSb	532, 2.33 473, 2.62 405, 3.06	~4 ~11 ~25	1 + 1.1	0.38 0.58 0.80	0.56 ± 0.03 0.69 ± 0.03 0.87 ± 0.04
GaAs(Cs,F)	532, 2.33	~10	1.4 ± 0.1	0.77	0.47 ± 0.03
GaN(Cs)	260, 4.77	~15	3.4 ± 0.1	0.94	1.35 ± 0.11

To analyze the photoemissive properties of semiconductor cathodes, we must consider various factors that influence the photoemission from their surfaces. In previous discussions we have considered the work function of a material as a bulk property determined by the electronic density of states within the solid, assuming an idealized, infinite semiconductor. For real applications, however, we must consider the interface between the semiconductor and the surrounding medium. At the surface, the abrupt termination of the lattice periodicity leads to a redistribution of charges, creating electronic energy states within the bandgap [43]. In case of intrinsic semiconductors, no band bending can be observed due to lattice termination, since the Fermi level is exactly in the middle of the bandgap, therefore no charge redistribution occurs. In case of n-type semiconductors, electrons will migrate from the bulk to the surface, causing the band to bend upwards. In p-type semiconductors, electrons will travel from the surface to the bulk, causing the band to bend downward [44]. Photoemission is generally more efficient in p-doped semiconductors because the downward band bending promotes electron escape [36]. Common examples of p-type semiconductor photocathodes are Cs₂Te [45], Na₂KSb [46] or GaAs [47]. The band bending effects due to surface states are illustrated in Figure 3.2.

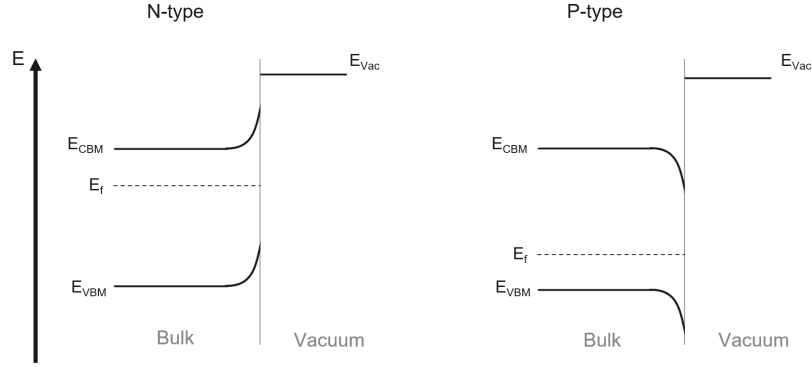


FIGURE 3.2: Surface state induced band bending in n-type and p-type semiconductors (Figure modified from [44]). The energy levels shown include the conduction band minimum (E_{CBM}), the valence band maximum (E_{VBM}), the Fermi level (E_f), and the vacuum level (E_{Vac}).

Photocathodes can be further divided into positive electron affinity (PEA) and negative electron affinity (NEA) types. Alkali-based photocathodes—such as Cs_2Te , Cs_3Sb , Na_2KSb , and K_2CsSb —are examples of PEA photocathodes, while materials such as GaAs and GaN are considered NEA photocathodes [10]. In an NEA surface state, the vacuum level E_{Vac} lies below the conduction band minimum E_{CBM} , making it easier for electrons to escape with less energy input. On the other hand, in PEA surface states, the E_{Vac} lies above the E_{CBM} , so the escaping electron faces a higher energy barrier to escape. This additional hurdle results in more energy being required for photoemission [36]. The band structures for both PEA and NEA semiconductors are shown in Figure 3.3.

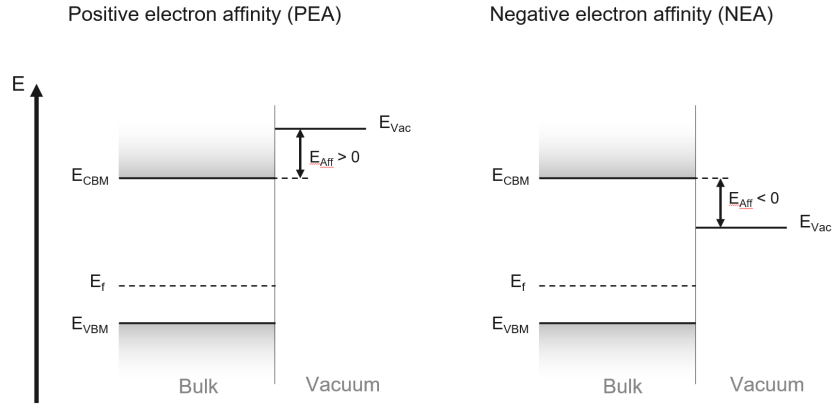


FIGURE 3.3: Comparison of PEA and NEA cathodes showing the location of the vacuum level relative to the conduction band minimum (Figure modified from [48]). The energy levels shown include the conduction band minimum (E_{CBM}), the valence band maximum (E_{VBM}), the Fermi level (E_F), the vacuum level (E_{vac}), and the electron affinity (E_{Aff}).

In general, NEA cathodes are advantageous for photoemission because they require less energy for electrons to overcome the barrier. However, photoemission in PEA-type semiconductors can be enhanced by surface engineering to lower the energy barrier. This can be achieved by depositing a thin layer of a low work function material, such as an alkali metal, to change the surface properties and lower the vacuum energy level [36]. For example, research by Ghosh et al. has shown that adding cesium and antimony to Na_2KSb , a heavily p-doped material, reduces the electron affinity from 0.7 eV to 0.24 eV, effectively creating an NEA surface state [46]. In this way it is possible to combine in a cathode properties that are advantageous for photoemission: p-doping and an NEA surface state.

III-V semiconductors (GaAs and GaN) owe their good photoemission properties to NEA conditions established on their surface. This happens through the deposition of a highly electropositive alkali metal, such as Li, Na, K, Rb, or Cs, with Cs being the most electropositive [10]. Figure 3.4 shows the electronegativity trends of the periodic table.

Electronegativity refers to the tendency of an element to attract electrons in a chemical bond. The more electronegative the element, the more it attracts electrons. Conversely, when a highly electropositive element is deposited on the surface, it attracts electrons less strongly, creating a positive dipole moment at the surface that produces NEA conditions. An example of NEA activation in GaAs photocathodes is achieved by

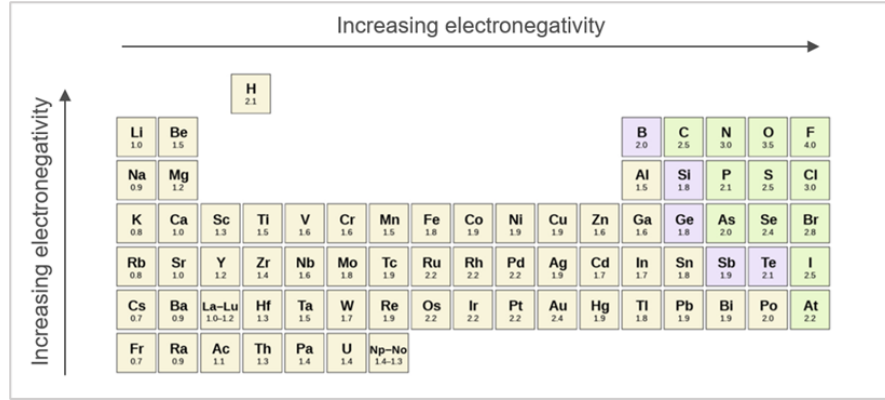


FIGURE 3.4: Electronegativity trends of the periodic table [49].

cesium and oxygen adsorption, which forms a Ga-O-Cs dipole on the surface, thereby lowering the E_{Vac} barrier [36, 50]. For GaAs, it has also been shown that the E_{Vac} barrier can also be lowered by Cs-Te deposition, as shown in Figure 3.5 [47].

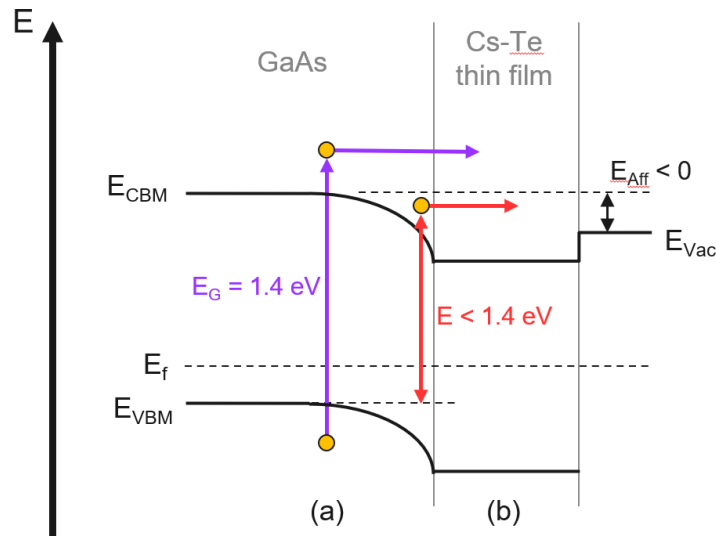


FIGURE 3.5: Photoemission from (a) a p-type semiconductor with downward bending surface state and (b) lowered E_{Vac} due to NEA conditions, achieved by depositing a thin Cs-Te film (Figure modified from [47]). The energy levels shown include the conduction band minimum (E_{CBM}), the valence band maximum (E_{VBM}), the Fermi level (E_f), the vacuum level (E_{Vac}), and the electron affinity (E_{Aff}).

3.2 Photocathode Fabrication

At CERN, photocathodes can be fabricated in situ in the vacuum chamber of the photoinjector, such as in the CLEAR facility, or fabricated externally in the CERN Photoemission Laboratory and then transported to the photoinjector or RF gun using a UHV "suitcase" (transport carrier). A schematic layout of CERN's Photoemission Laboratory setup is shown in Figure 3.6.

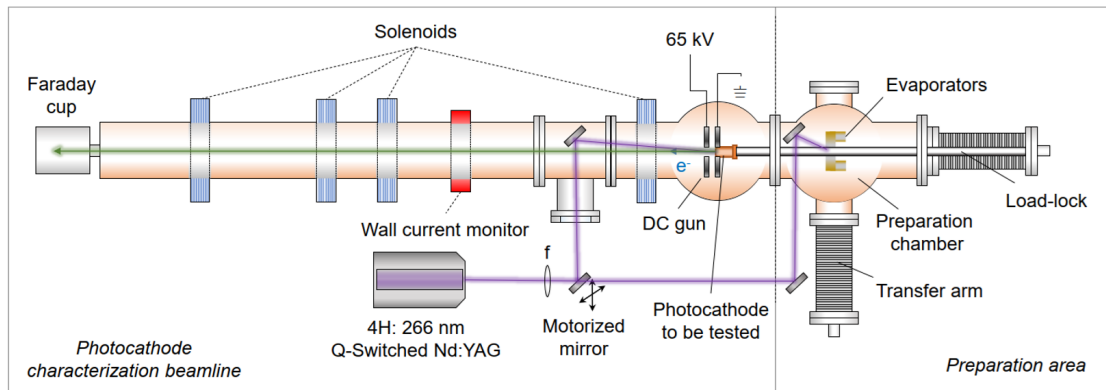


FIGURE 3.6: Schematic layout of CERN's Photoemission Laboratory setup [51].

The photocathode production process begins in the preparation chamber, where photocathodes are coated with a photoemissive material. At CERN, Cs-Te is currently used due to its superior properties, as described in Chapter 3.1 *Photocathode Materials*. However, the evaporator setup is configurable to allow the use of other materials and can contain individual sources for elements such as Cs, Te, Sb, K, Ga and As, allowing flexible combinations of compounds. A 3D model of the evaporator setup is shown in Figure 3.7.

Once the photocathode is in the preparation chamber, a thin film of Cs-Te is deposited on a copper plug, using physical vapor deposition (PVD). Since Cs-Te is highly reactive and sensitive to impurities and contamination by residual gases, it is crucial that all areas involved in the production and handling of cathodes remain under ultra-high vacuum (UHV) conditions (10^{-11} mbar). The Transport Carrier is used to introduce new photocathodes by venting, disconnecting, and opening it. After the insertion, it has to be reconnected and baked out. A UHV Transport Carrier with 4 slots is used to store and transport photocathodes between the Photoemission Lab and the AWAKE installation. These UHV transport vessels connect to the preparation chamber through

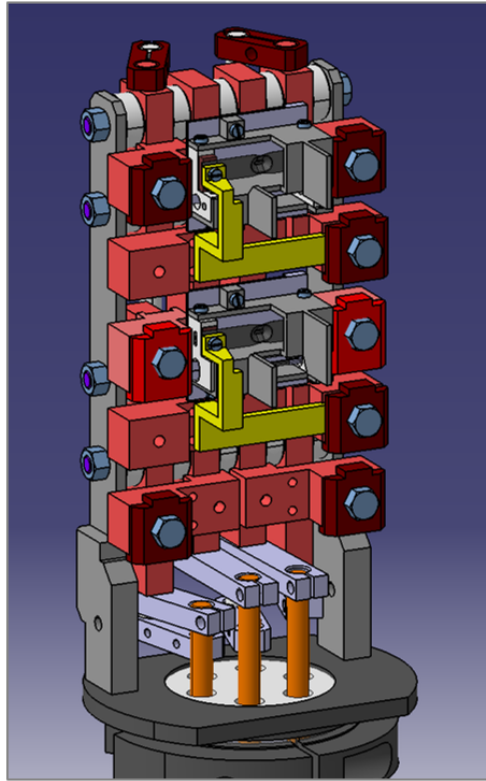


FIGURE 3.7: 3D Model of the Evaporator Setup in CERN's Photoemission Laboratory [21].

an intermediate section, requiring a bake-out operation after (re)attachment to reach the required UHV level of 10^{-11} mbar before the section valves are opened. Two integrated horizontal manipulators work together to select, move and place the desired photocathodes in the vacuum chambers.

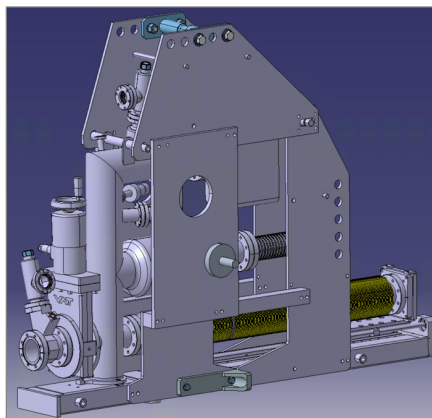


FIGURE 3.8: 3D Model of the UHV Transport Carrier in CERN's Photoemission Laboratory [21].

Once the production process is complete, the photocathode is transferred to the characterization beamline for testing. A load-lock system equipped with a photocathode manipulator facilitates the transfer of the photocathode between the two chambers, as shown in Figure 3.9.

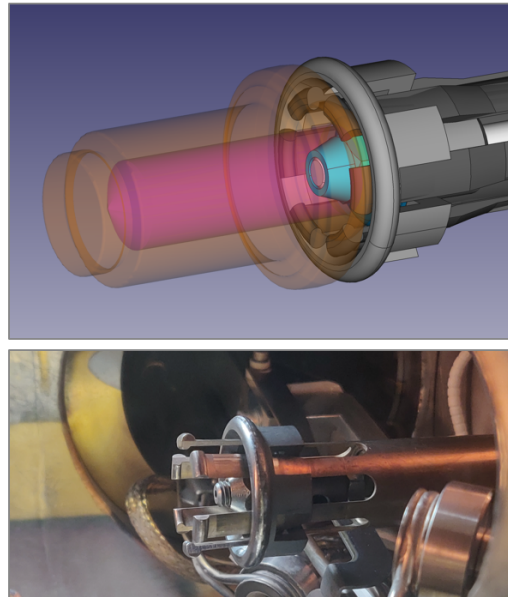


FIGURE 3.9: Photocathode manipulator used in the Photoemission Laboratory [21].

In the characterization beamline, the photocathode is tested in the DC electron gun. After the photocathode is inserted into the gun and the system is ramped up to 65 kV, beam guidance magnets (solenoids) and measurement devices are turned on and the characterization process can begin. Four instruments are installed to measure the electron beam current: a Faraday Cup (FC), a Wall Current Monitor (WCM), a Fast Current Transformer (FC), and a Cylinder (CY), enabling precise characterization of the photocathode quantum efficiency. The photoemission drive laser, a Nd:Yag laser operating at its fourth harmonic (266 nm), is used for standard testing and production monitoring. A motorized mirror allows the laser beam to be switched between the photocathode in the preparation chamber or in the gun.

The quality of the photocathode is highly dependent on the parameters used in the coating process. Coating is achieved by physical vapor deposition, a thin film deposition technique in which the desired material is evaporated by physical means and then condensed onto a substrate to form a thin film [52]. The evaporation process can be achieved by various methods, the most common of which is thermal evaporation.

Heating techniques such as resistive heating, inductive heating, or electron beam heating are used to achieve the temperatures required for evaporation [53].

For many thin film applications, it is critical to achieve the correct stoichiometric ratio of materials during the PVD process. In the case of molecular solids, their vapor typically consists of the desired stoichiometric composition, allowing direct evaporation. However, this is not universally applicable; some materials must be evaporated separately from their constituents, making stoichiometric control difficult. For semiconductor thin film deposition, the co-evaporation method using a three-temperature approach (see Figure 3.10) has proved effective in accurately controlling the deposition composition [52].

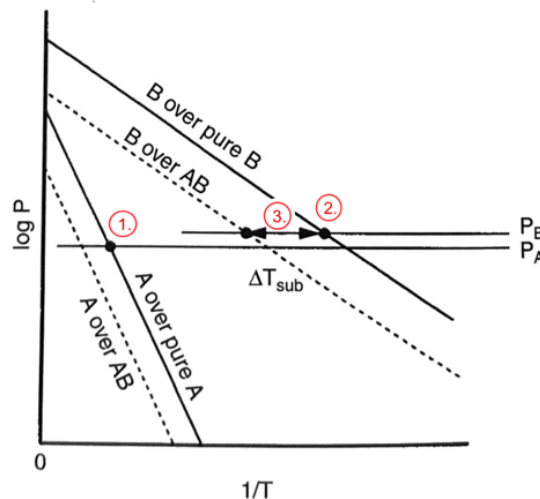


FIGURE 3.10: Phase diagram illustrating the three-temperature method for physical vapor deposition (PVD) [52].

For binary compounds, optimal deposition temperature parameters can be determined from the phase diagram of the individual components, as the vapor pressure directly affects the rate of evaporation and subsequent deposition. In the initial step (labeled as Step 1 in Figure 3.10), the vapor pressure of the less volatile component (Component A) is selected to achieve a reasonable deposition flux, which determines the deposition rate of the compound (AB). Next, the vapor pressure of the more volatile component (Component B) is selected (Step 2 in Figure 3.10). To ensure complete reaction of Component A, the deposition rate of Component B is set about 10% higher than the stoichiometric requirement [52]. For Cs-Te cathodes produced at CERN, tellurium

(Te) is the less volatile component, while cesium (Cs) is the more volatile one; their respective boiling points are listed in Table 3.2.

TABLE 3.2: Boiling Points of Cesium and Tellurium.

Element	Boiling Point	Reference
Cs	671 °C	[54]
Te	988 °C	[55]

Once the vapor pressures and therefore the evaporation temperatures for both components have been determined, the substrate temperature at which the compound will condense must be selected (Step 3. in Figure 3.10). This temperature should fall within the condensation window, shown as ΔT_{sub} in Figure 3.10. The upper limit of this window is defined by the 'B over AB' line; at temperatures above this limit, component B will remain in the gas phase and will not condense. The lower limit is defined by the 'B over pure B' line, where lower temperatures would lead to the formation of a two-phase film of B and AB. Therefore, the substrate is often heated to stay within this window [52]. In the literature, some procedures recommend heating the substrate up to 120 °C for Cs-Te deposition [56]. However, in the Photoemission Laboratory at CERN, the copper substrate is left unheated during deposition, as previous experiments with substrate heating have not yielded superior results.

To ensure the correct stoichiometric deposition ratio during co-evaporation of Cs and Te, the thickness ratio η of the deposited materials can be estimated using the following formula [51]:

$$\eta = \frac{\rho(\text{Cs})t(\text{Cs})m(\text{Te})}{\rho(\text{Te})t(\text{Te})m(\text{Cs})}, \quad (3.1)$$

where ρ is the density of the respective component, t is the deposited thickness measured by the microbalances, and m is the atomic mass. The constants needed for these calculations are listed in Table 3.3.

TABLE 3.3: Constants for Calculating the Stoichiometric Deposition Ratio of a Cs-Te film [54, 55].

Element	Density (ρ)	Atomic Mass (m)
Cs	1.90 g/cm ³	132.9 u
Te	6.23 g/cm ³	127.6 u

To achieve reproducibility in the deposition process, it is essential to control not only the stoichiometry but also other deposition conditions that affect the thin film properties. These factors include substrate surface conditions (e.g. roughness, surface flaws and contaminants), as well as the angle of incidence during deposition, growth morphology, preferred nucleation sites, gas entrapment and potential chemical reactions between the film and the substrate. Thin film deposition is therefore a complex process and can produce films with unique properties depending on the deposition parameters [57].

Thermal evaporation is generally performed under high vacuum conditions (10^{-6} to 10^{-7} mbar or better) to allow the evaporated material to reach the substrate with minimal collisions, ensuring that the mean free path is greater than the distance between the source and the substrate [53]. In addition, maintaining UHV conditions of 10^{-11} mbar [51] is critical to prevent degradation of the photocathode material, particularly as Cs is highly reactive [54].

Two main methods are used to thermally evaporate and deposit Cs and Te onto the uncoated photocathode surface: sequential and co-deposition [51]. Sequential deposition is the most widely established method for fabricating Cs-Te photocathodes. The procedure, as proposed by Di Bona et al. [56], involves first depositing a 10 nm layer of Te onto the substrate, followed by a Cs layer. The thickness of the Cs layer is determined experimentally by monitoring the photocurrent generated by a laser, and the Cs dispenser is turned off once the photocurrent reaches its maximum value [58, 59]. During this process, two quartz microbalances are used to monitor the thickness of the deposited layers. Each material, Cs and Te, has its own microbalance, which are partially shielded from each other to allow independent monitoring of the film thickness (see Figure 3.11). The dispensers for Cs and Te are indirectly heated using resistive heating, which is achieved by passing a current flow through the material to allow evaporation of Cs and Te [51].

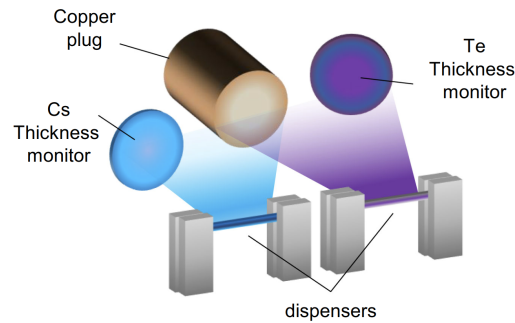


FIGURE 3.11: Schematic layout of the deposition process, illustrating the arrangement of the Cs and Te dispensers along with their respective thickness monitors [51].

In contrast to the sequential deposition process, the co-evaporation process does not require the initial deposition of a Te layer. Instead, Cs and Te are simultaneously vaporized and mixed in the gas phase before being deposited on the substrate. This method, originally proposed by G. Ghost and B. P. Varma [46] for alkali-antimony cathodes, has been tested at CERN's Photoemission Lab for Cs-Te photocathodes.

Research by G. Suberlucq [60] at the Photoemission Laboratory of CERN demonstrated that co-evaporation leads to longer cathode lifetimes and higher quantum yields compared to sequential deposition. The photocathodes were tested at a laser wavelength of 262 nm, an operating pressure of $< 10^{-9}$ mbar, and an electric field of 127 MV/m. During these tests, a single extracted charge per pulse of 100 nC was achieved within a pulse duration of 10 ps. The work conducted on the lifetimes of sequential and co-deposited cathodes is illustrated in Figure 3.12.

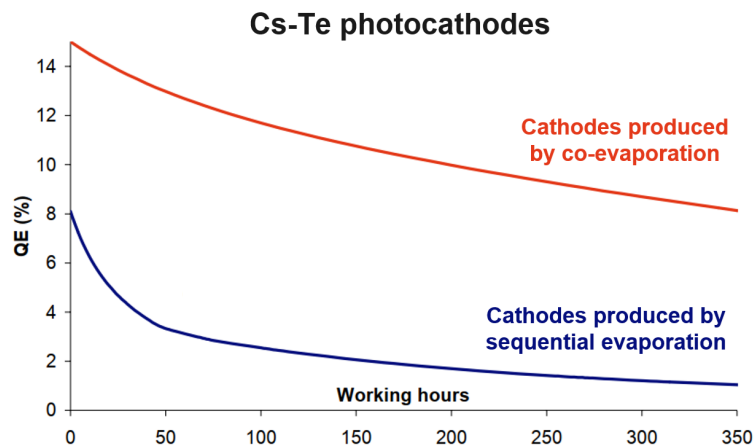


FIGURE 3.12: Lifetime of photocathodes fabricated by sequential and co-deposition [60].

Further studies, as reported by E. Chevallay [19], focused on monitoring thin film growth alongside the corresponding stoichiometric deposition ratio and quantum efficiency. Cathodes grown with Cs-Te deposition ratios between 2:1 and 2.5:1 achieved quantum efficiencies of 16-18%, at which point the growth process was stopped.

The photocathode fabrication process can be performed on a variety of substrates, with copper and molybdenum being the most commonly used materials. At CERN, a 19 mm diameter polished electronic grade copper plug is typically used as the substrate [51].

3.3 Effects of Cathode Impurities

During the production of Cs₂Te photocathodes, other stoichiometric compounds of cesium telluride (Cs-Te) or pure cesium (Cs) may also condense on the surface. As discussed in the previous chapter, a thin layer of cesium effectively lowers the work function of materials with inherently higher work functions. However, precise control of the deposition process for compounds that are not molecular solids is challenging and can lead to uneven or excessive deposition of one or both components. At CERN's Photoemission Laboratory, our focus is on Cs₂Te photocathodes deposited on a copper plug.

According to the literature, pure Cs has a work function of approximately 2.14 eV, whereas copper has a work function of around 4.7 eV. For Cs₂Te, reported values indicate a bandgap energy of 3.3 eV and an electron affinity of approximately 0.2 eV. Excess Cs on the photocathode surface can lower the bandgap of Cs₂Te and reduce the vacuum energy barrier. Applying a thin film of a low-work-function material like Cs can lower this barrier, which is beneficial for increasing photoemission probability in some applications. However, Cs₂Te is known for its high photoemission quantum yield, and achieving optimal photoemission requires matching the photon wavelength closely to its bandgap [10]. If the energy barrier is lowered due to excess Cs or other impurities, the resulting lower barrier can increase the excess kinetic energy of the emitted photoelectrons, potentially affecting photoemission yield. The effect of overcesiation on Cs₂Te photocathodes is illustrated in Figure 3.13.

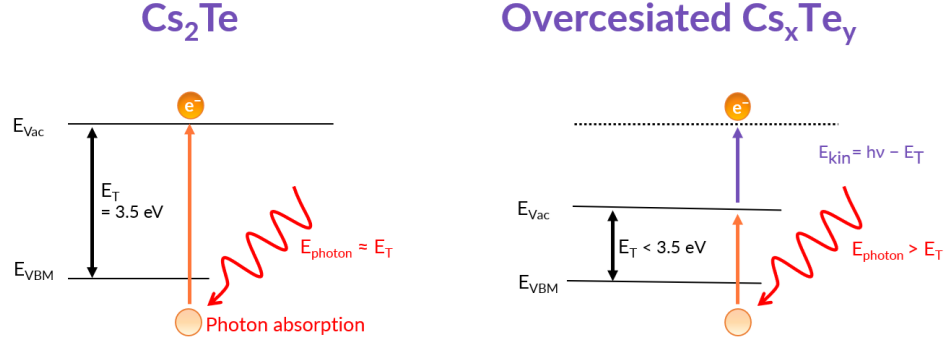


FIGURE 3.13: Photoemission scheme for Cs_2Te and for overcesiated Cs-Te compounds, demonstrating the bandgap lowering effects of cesium.

The excess kinetic energy of emitted electrons can be written as follows [61]:

For metals:

$$E_{\text{kin, metals}} = h\nu - \Phi \quad (3.2)$$

For semiconductors:

$$E_{\text{kin, semiconductors}} = h\nu - E_T \quad (3.3)$$

Below the threshold energy, photoemission does not occur. When photon energy exceeds this threshold, however, the surplus energy is converted into the kinetic energy of the emitted electrons. As this kinetic energy increases, so does the transverse momentum component $p_{\perp}$ of the emitted electrons [62], leading to a higher momentum spread of the particles and thus a higher Mean Transverse Energy (MTE) [63]

$$\text{MTE} = \frac{E_{\text{kin}}}{3} = \frac{p_{\perp}^2}{2m} . \quad (3.4)$$

This relationship influences the variance of the transverse momentum $\sigma_{p_x}^2$ and consequently the dimensionless transverse momentum spread σ_{p_x} of the beam [17]

$$\sigma_{p_x} = \sqrt{\frac{\text{MTE}}{mc^2}} . \quad (3.5)$$

This affects the normalized transverse emittance $\varepsilon_{n,x}$ of the beam [63]

$$\varepsilon_{n,x} = \sigma_x \sigma_{p_x} , \quad (3.6)$$

where $\epsilon_{n,x}$ is the root-mean-square (rms) normalized emittance, representing the spread of particle positions and momenta within the beam and σ_x is the rms laser spot size on the cathode. In Figure 3.14, a schematic diagram illustrates how an increase in the kinetic energy of emitted electrons affects the mean transverse energy (MTE) of the beam, which affects the emittance of the beam.

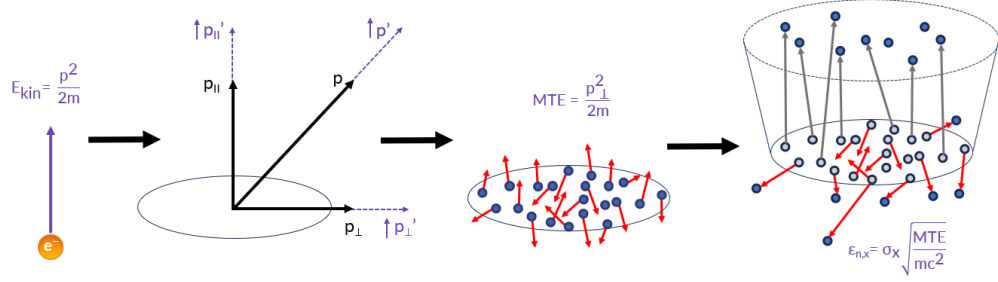


FIGURE 3.14: Schematic representation of the effects of high electron kinetic energy on beam dynamics.

Finally, this relationship directly affects beam brightness [64]

$$B = \frac{2I}{\pi^2 \epsilon_{n,x} \epsilon_{n,y}} \left[\frac{\text{A}}{\text{m}^2 \text{rad}^2} \right], \quad (3.7)$$

where I is the peak current and $\epsilon_{n,x}$, $\epsilon_{n,y}$ are the normalized emittances of the x and y components, respectively. Beam brightness is a measure of the electron volume density in the 6-dimensional phase space. In this space, x , y , and z represent position coordinates, while x' , y' , and z' represent angular directions determined by the particles' momenta. [10]

In case of Cs_2Te , it is known that it has a bandgap of 3.3 eV and an E_A of 0.2 eV. A high E_G/E_A ratio, as seen in Cs_2Te , is known to have a positive effect on quantum efficiency, increasing the likelihood of electron escape with minimal energy loss [45]. For Cs, it is known from the literature to have a work function of 2.14 eV [65].

If additional Cs is deposited on the Cs_2Te surface, it can modify the photoemission properties, lowering the threshold energy. For pure Cs_2Te , the photoemission threshold is expected to be around 3.5 eV. In work from the 1950s, Taft and Apker [66] used spectral emittance measurements to reveal that when excess cesium was present on the Cs_2Te surface, a low-energy shoulder appeared in the emission spectrum. This shoulder indicated that photoelectrons could emit at energies below the expected 3.5

eV threshold, which they attributed to the influence of excess Cs lowering the energy barrier.

In the 1970s, Powell et al. revisited this phenomenon and proposed that the low-energy shoulder was not simply due to excess Cs but likely resulted from the formation of a different Cs_xTe_y phase, where $x \neq 2$. Their experiments aimed to test this hypothesis by adding additional tellurium and heating the Cs_2Te surface, which was expected to convert these alternative phases back to pure Cs_2Te and thereby eliminate the low-energy shoulder in the emission spectrum. While the shoulder did decrease, it was not completely eliminated, suggesting that multiple Cs_xTe phases might coexist on the surface, each influencing the photoemission properties. This finding highlights the complexity of surface composition and phase structure in determining the photoemission threshold of Cs_2Te photocathodes [59, 67]. The spectral responses observed in the studies by Taft and Apker, as well as Powell et al., are illustrated in Figure 3.15.

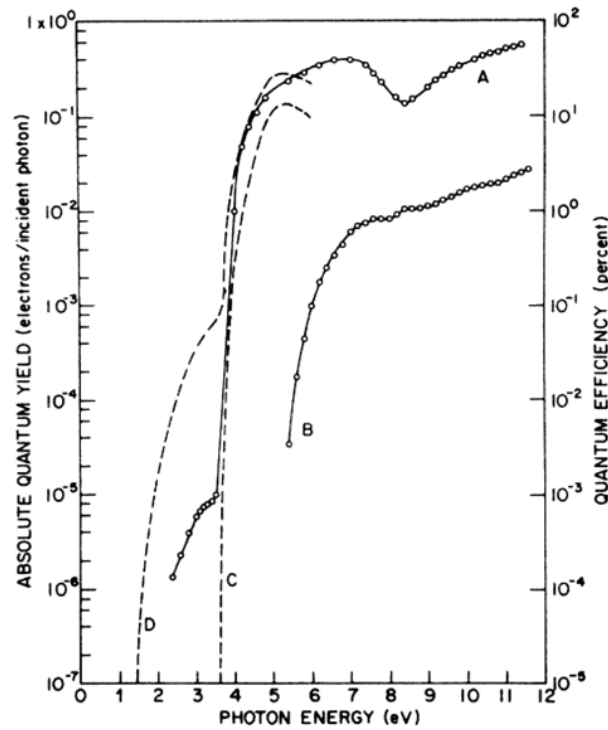


FIGURE 3.15: Spectral distribution of the quantum efficiency. Curves (a) and (b) are from Powell et al. [67], where (a) represents pure Cs_2Te and (b) shows evaporated Te before any reaction with Cs. Curves (c) and (d) are from Taft and Apker [66], with curve (c) illustrating pure Cs_2Te and curve (d) showing Cs_2Te after extended exposure to Cs.

Since these studies, it has been historically confirmed that the low-energy shoulder is most likely attributed to the presence of metallic Cs and the compound Cs_5Te_3 . The existence of Cs_5Te_3 has been demonstrated using both X-ray Photoelectron Spectroscopy (XPS) and X-ray Diffraction (XRD). Cs_5Te_3 has a low bandgap of 1.15–1.20 eV, and cesium itself has an inherently low work function of 2.14 eV. Together, these properties make it easier for electrons to be emitted at lower photon energies, explaining the low-energy shoulder in the spectral response. The effects of overcesiation were further investigated by Pierce et al. by creating cesium-rich and low-cesium samples. The results showed the presence of a distinct shoulder below 3.5 eV only in the cesium-rich samples, as shown in Figure 3.16 [59].

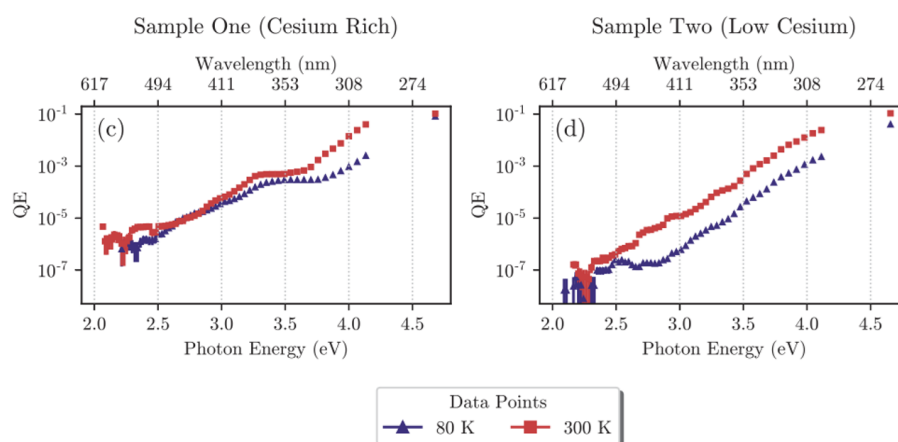


FIGURE 3.16: Spectral response of cesium-rich and low-cesium Cs-Te photocathodes as studied by Pierce et al. [59].

In another study conducted by Jones et al. [68], the effects of oxygen exposure were examined in the low-energy range. During the manufacturing process of Cs_2Te , it is unavoidable that elemental cesium gets produced, which then readily oxidizes when exposed to oxygen to Cs_2O . This compound has a low work function of 1.4 eV, and it has been concluded that the work function reduction and thus the low-energy shoulder is strongly related to the level of oxygen exposure. The spectral response measurement from their work is shown in Figure 3.17, together with the Cs:Te stoichiometric ratio from their production data in Table 3.4.

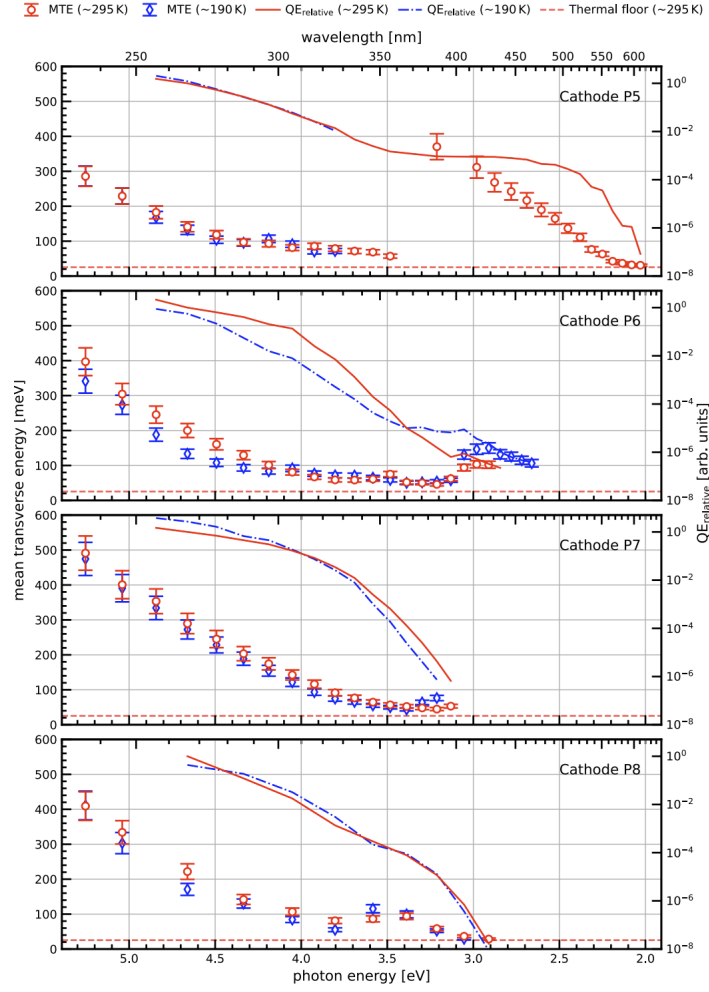


FIGURE 3.17: Photoemission spectrum of four Cs-Te photocathodes made at CERN, analyzed by Jones et al. [68].

TABLE 3.4: Final elemental stoichiometric deposition ratio of the samples (P5-P8) [68].

Sample	Cs:Te Ratio
P5	2.9 : 1
P6	2.6 : 1
P7	2.8 : 1
P8	1.5 : 1

The spectral response analysis shows a pronounced low-energy shoulder below 3.5 eV for cathode P5, the most cesium-rich sample. This shoulder was attributed to elemental cesium and cesium oxide (Cs_2O). In contrast, cathodes P6 and P7, which had less excess cesium according to the production data, did not show the pronounced shoulder

as seen in P5. However, both P6 and P7 showed evidence of oxygen poisoning on their surfaces, with P6 also containing carbon. XPS data indicated that cesium and tellurium in P6 and P7 were fully bonded, consistent with the photoemission spectrum showing a single photoemissive phase. Their threshold was recorded below 3.5 eV, likely due to a cesium-oxide phase. Cathode P8 showed no chemical poisoning by oxygen, and cesium was in fully bonded form according to the XPS data. A light shoulder appeared in its spectral analysis but was not further investigated, probably due to its minimal prominence. These results suggest that photoemission below the threshold energy of Cs_2Te can be greatly enhanced by the presence of elemental Cs and Cs_2O [68].

3.4 Rejuvenated Photocathodes

Cs-Te photocathodes are chemically highly reactive, which limits their lifetime and requires frequent intervention to replace or rejuvenate the Cs-Te film. Rejuvenated photocathodes offer a potential solution for quicker replacement in photoinjectors by allowing a new Cs-Te layer to be deposited over a degraded one, possibly without opening the vacuum chamber if done in situ. A common method of rejuvenation is to reactivate the Cs layer by heating it to 120-200 °C for 1-3 days while simultaneously exposing it to UV light. According to the literature, this method can recover approximately 60% of the original QE [56, 69], as the Cs gets redistributed on the surface [51]. Another rejuvenation approach is to deposit a new thin film over the degraded one. Two techniques can be considered: sequential deposition and co-deposition. A comparison of the sequential and co-deposition rejuvenation strategies is shown in Figure 3.18.

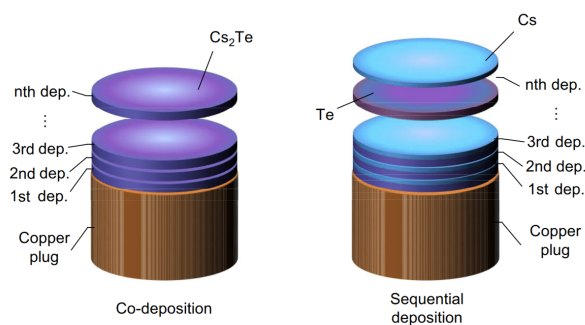


FIGURE 3.18: Schematic layout of the deposition strategy for co- and sequential deposition [51].

At CERN, sequential deposition has been routinely practised at the CLEAR experiment. In this process, multiple Cs-Te layers are deposited over the degraded cathode with 10 nm of tellurium and 40 nm of cesium in situ at gun level without breaking the UHV conditions. Figure 3.19 illustrates the QE history of photocathode rejuvenation at CERN's CLEAR facility from 2018 to 2024 [51].

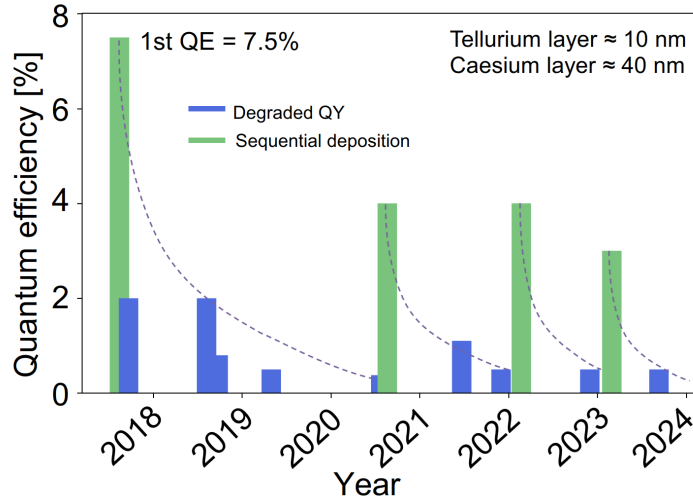


FIGURE 3.19: QE evolution of photocathodes rejuvenated by sequential deposition used in CERN's CLEAR facility [51].

Figure 3.19 shows that a QE of approximately 7.5% was achieved with the first deposition on the AWAKE cathode. Subsequent rejuvenation processes resulted in a QE of 3-4%, as shown in Table 3.5.

TABLE 3.5: QE values for photocathode rejuvenation by sequential deposition at CERN's CLEAR facility [51]

Sequential deposition	QE [%]	Electron gun type
First deposition (2018)	7.5	RF gun
First rejuvenation (2021)	~4	RF gun
Second rejuvenation (2022)	~4	RF gun
Third rejuvenation (2023)	~3	RF gun

Another approach to rejuvenation is co-deposition, where a new co-deposited Cs-Te layer is applied over a degraded one. This method was first proposed and investigated by Martinez-Calderon et al., who demonstrated its effectiveness by evaluating the quantum efficiency (QE) of two rejuvenated cathodes. They performed surface scans to generate QE maps for both the fresh and by co-deposition rejuvenated cathodes, allowing a clear comparison, as shown in Figure 3.20 [51].

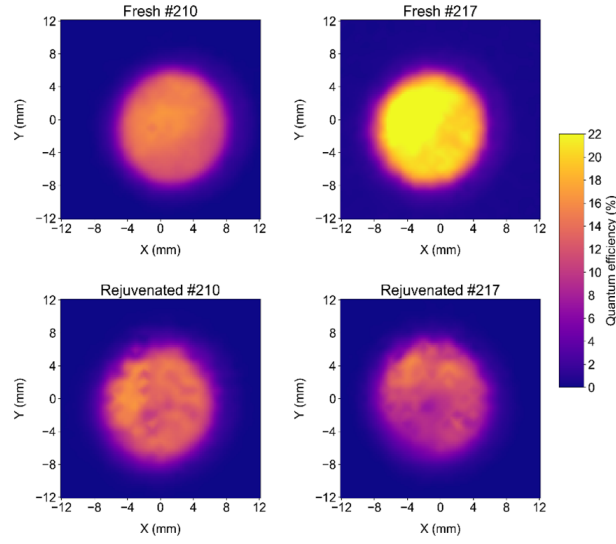


FIGURE 3.20: QE surface profile of fresh and by co-deposition rejuvenated cathodes at CERN [51].

Figure 3.20 shows that the rejuvenated cathodes exhibited a significantly higher spatial inhomogeneity in QE compared to their states prior to rejuvenation. The QE maps were obtained using a 10 Hz repetition rate laser at 266 nm [51]. Table 3.6 shows the measured QE values for the two fresh cathodes, together with their degraded QE values and the corresponding freshly rejuvenated QE values.

TABLE 3.6: QE values for photocathode rejuvenation by co-deposition at CERN [51]

Cathode	Co-deposition	QE [%]	Electron gun type
#210	Fresh	16 / 12	Dc gun / rf gun
	Degraded	0.16	Rf gun
	Rejuvenated	15	Rf gun
#217	Fresh	25 / 23	Dc gun / rf gun
	Degraded	0.01	Rf gun
	Rejuvenated	14	Rf gun

After their first fabrication, both photocathodes were stored for about four years before being used in the AWAKE experiment at CERN. Photocathode #210 had an initial QE of 16%, with a stoichiometric deposition ratio, calculated using Equation 3.1, of 3.91 based on the total thickness of Cs and Te. The cathode was used in the AWAKE experiment for two years before being placed in a transport carrier for storage until rejuvenation. Prior to rejuvenation, the QE value was recorded as 0.16%, probably due to chemical poisoning or laser damage to its surface [51].

In the case of photocathode #217, it had an initial QE of 25% with a stoichiometric deposition ratio of 3.93. After nine months of operation, it was accidentally exposed to low vacuum conditions, which caused its QE to drop to 0.01%. This decline can be attributed to oxygen exposure during the low vacuum incident [51]. The fabrication data for the two fresh photocathodes are shown in Figure 3.21.

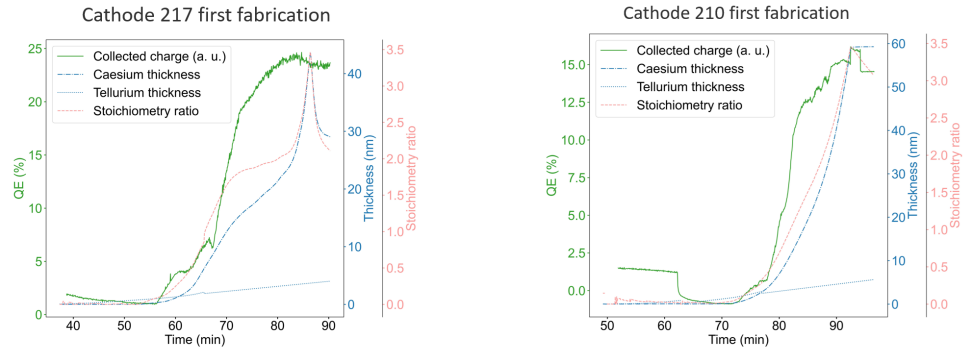


FIGURE 3.21: First fabrication data for cathodes #210 and #217 [51].

Following degradation, both cathodes were rejuvenated by co-deposition in the photoemission laboratory. Photocathode #210 achieved a final QE of 15%, representing a recovery of 95% of its original QE, with an overall stoichiometric deposition ratio of 1.24. Meanwhile, cathode #217 achieved a final QE of 14%, representing a recovery of 56% of its initial QE, with a final stoichiometric deposition ratio of 3.27 [51]. The fabrication data for these rejuvenation processes are shown in Figure 3.22.

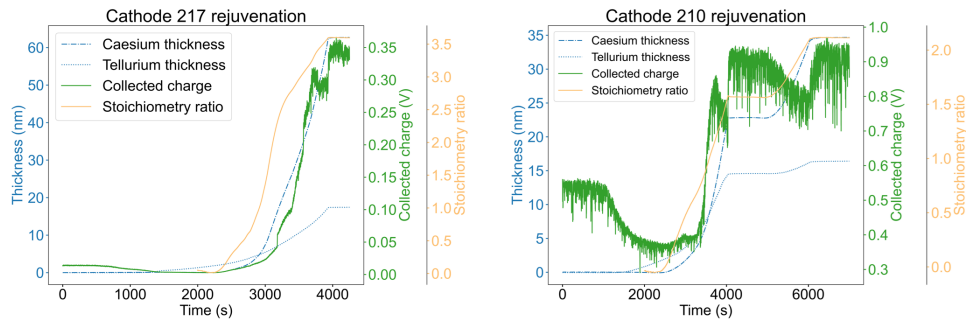


FIGURE 3.22: Rejuvenation fabrication data for cathodes #210 and #217 [51].

TABLE 3.7: Stoichiometric deposition ratios for photocathodes #210 and #217 [51].

Cathode	Fresh Ratio	Rejuvenated Ratio
#210	3.91	1.24
#217	3.93	3.27

Chapter 4

Experimental Setup

This study focuses on rejuvenated photocathodes, which are considered for their potential to serve as a non-disruptive and efficient option for quick photocathode replacement in photoinjectors. Previous chapters established that Cs_2Te , the photoemitting material currently used at CERN, offers high quantum efficiency and greater durability compared to alternative semiconductor materials. To achieve optimal photoemission, the photon energy of the driving laser must match the specific photoemission threshold of the material. However, as discussed in Chapter 3.3 *Effects of Cathode Impurities*, controlling the deposition of stoichiometric compounds such as Cs-Te poses significant challenges, often resulting in excess Cs being incorporated into the film. Given the inherently low work function of Cs, this excess Cs reduces the threshold energy required for photoemission, resulting in higher kinetic energy of the emitted electrons, which in turn increases beam emittance and reduces beam brightness.

To evaluate the quality of photocathodes generated by rejuvenation via co-deposition, a spectral response analysis was carried out. By scanning the laser wavelength near the photoemission threshold and then record the quantum efficiency as a function of photon energy, this analysis allows for an approximation of the threshold energy for the coating material present on the photocathode. As a laser source, an optical parametric oscillator (OPO) was used. The resulting data provides valuable insights into the surface composition and photoemissive properties of the photocathode material. The primary objective of this investigation is to correlate spectral response data with the cathode fabrication parameters to potentially improve the photocathode fabrication process.

An improved photocathode could eliminate the low energy shoulder on the spectral response, thereby reducing the kinetic energy of the emitted electrons. This in turn would reduce the MTE of the electron beam, thus reducing beam emittance and increasing beam brightness, which are key parameters for high average current applications requiring precise electron bunches. For the spectral response analysis, the output of the OPO was directed onto the photocathode surface with a wavelength range of 240 to 430 nm, covering wavelengths near the photoemission threshold energy for Cs₂Te (approximately 3.5 eV).

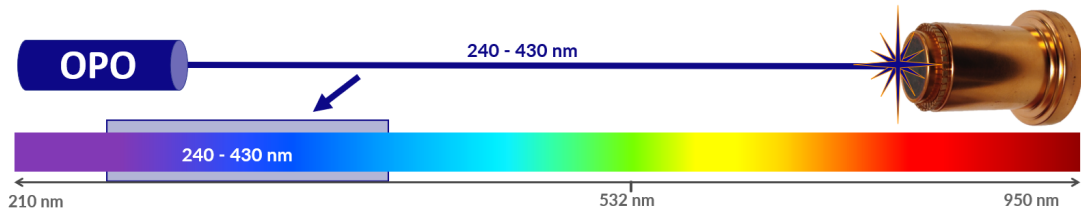


FIGURE 4.1: Schematic representation of the experimental setup.

4.1 Laser System

For this analysis, the optical table was set up using the NT230 Optical Parametric Oscillator (OPO) model from EKSPLA [70]. The OPO operates at a repetition rate of 10 Hz with a pulse duration of 5 ns. This system has multiple output channels, each designed for specific wavelength ranges. The first channel, called SCU (spectral cleaning unit), offers a wide tuning range from 210 to 2600 nm. The second output provides fixed wavelengths including 1064 nm (fundamental, H), 532 nm (second harmonic, SH), and 355 nm (third harmonic, TH). A third output provides SH, SF (sum frequency), and OPO outputs covering the wavelength range of 210 to 2600 nm. The fourth output, dedicated specifically to the OPO function, provides a wavelength range of 405 to 2500 nm. This tunable range is achieved by passing the laser beam through two non-linear beta-barium borate (BBO) crystals that generate different wavelengths through a process called parametric down-conversion, the basic principle of an OPO [71].

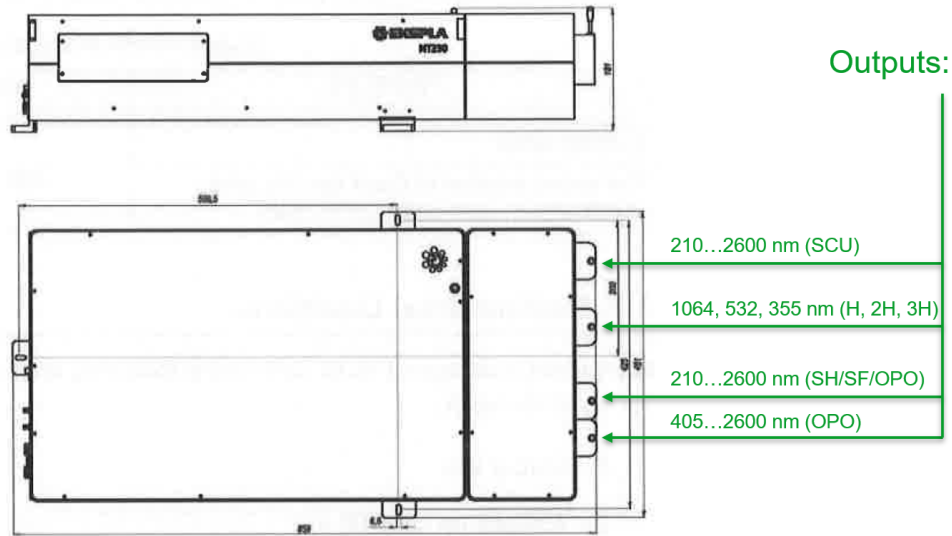


FIGURE 4.2: Outline of the NT230 laser series (Figure adapted from the EKSPLA NT230 manual).

Once the OPO is set up on the optical table, a spectrometer (Ocean Optics Flame, 200-1025 nm [72]) was installed to verify that the measured wavelength matched the wavelength specified for the OPO. Along the beam path, reflective filters with neutral densities (0.1, 0.2, 0.3, 0.5, 1, 1.3, 2 ND) [73] were installed to attenuate the laser intensity. In addition, a beam sampler was positioned to reflect one-third of the laser beam, creating a virtual beamline. This virtual beamline is designed to be the same length as the primary beamline, allowing monitoring of the beam shape and position as it reaches the cathode. The remaining beam was directed to a Gentec Maestro energy meter for accurate energy measurements, while UV-enhanced mirrors guided the beam along the path. The overall setup of the optical table for spectral response measurement is shown in Figure 4.3.

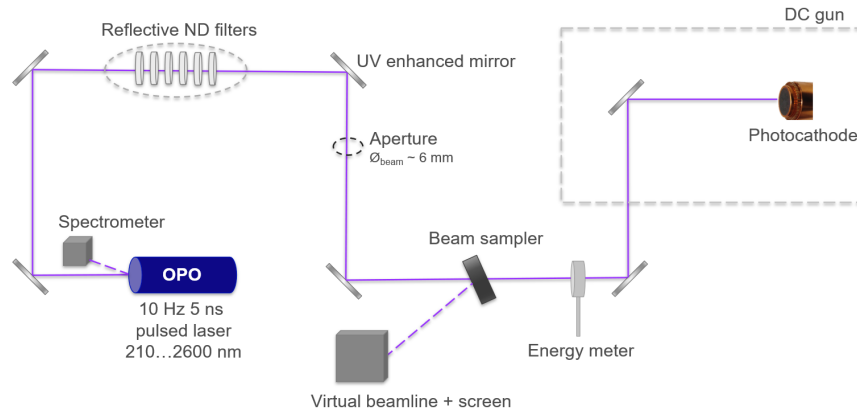


FIGURE 4.3: Optical table configuration for spectral response measurement.

Laser energy was measured using the Gentec sensor head QE8SP-B-MT-D0, with a sensitivity range of 80 nJ to 0.93 mJ [74]. The sensor was connected to the Gentec Maestro energy meter display, and the corresponding wavelength was manually set on the device. The laser beam was aligned with the energy meter head, and the system recorded the average energy over 100 laser shots. This average value was then used later to calculate the quantum efficiency. To validate the laser energy and measurement system, an initial test setup was implemented, as shown in Figure 4.4. This configuration ensured the reliability of the energy measurement components and procedures.

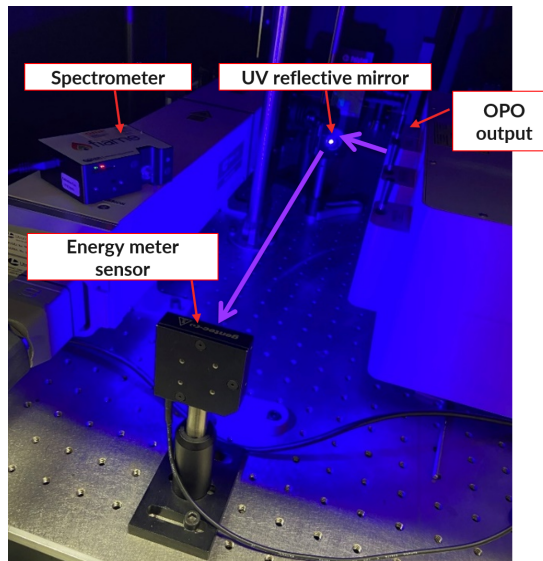


FIGURE 4.4: Initial configuration for laser energy measurement to test system components.

The energy output of the laser was characterized in the wavelength range of 200 to 530 nm and compared to the energy specifications in the OPO manual. To assess the attenuation characteristics of the UV-enhanced fused silica ND filters, the filters were first calibrated. Figure 4.5 shows a comparative plot of the specified laser output energies, the measured energies, and the energies after transmission through a single UV-enhanced 2ND filter. The OPO/SH/SF output was used for measurements in the 200-400 nm range and the OPO output was used for the 400-530 nm range. For more information on the output energy specifications of the OPO see Appendix A , and on the transmission values over a wider wavelength range for the UV-FS ND filters, see Appendix B.

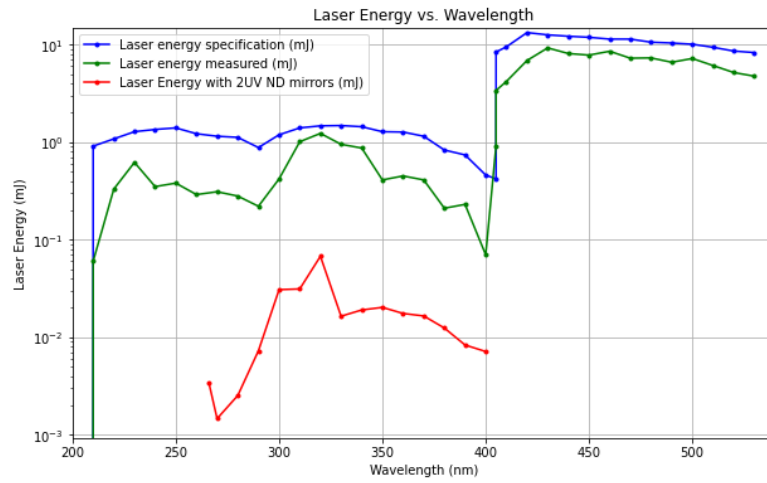


FIGURE 4.5: Laser energy characterization.

For successful beam alignment, the laser beam must pass through the last aperture, which requires manual beam alignment for each wavelength.

4.2 Electron Beamline

Once the laser beam is aligned and the machine is set up, the laser is directed onto the photocathode, inducing electron emission from the cathode surface. The resulting current is measured over time for each laser pulse. By measuring the electric potential (in volts), the corresponding charge (in coulomb) per pulse can be determined. To ensure that the extracted charge is captured within the measurement window, the

sync-out delay in the EKSPLA software was set to 177 μs , and a trigger delay of 955 ns was configured for the measurement equipment.

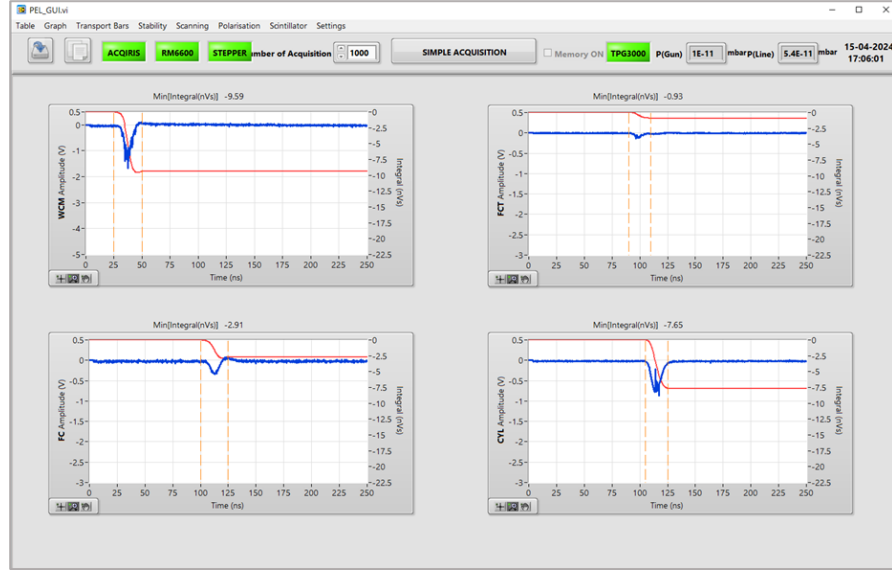


FIGURE 4.6: LabVIEW panel showing four measurement windows, each displaying the signal from different current measurement devices (WCM, FCT, FC, CLY).

To accurately measure the emitted charge, we must first center the cathode precisely in the machine. We begin by manually aligning the beamline using mirrors so that the profile of the sampled beam (virtual line) is centered on the screen, which can be verified on the optical table. Once aligned, we direct the laser onto the photocathode and use motorized mirrors to scan the beam in the XY-direction, creating a profile of the quantum efficiency across the cathode surface. During the scan, the XY-stage moves the beam across the cathode surface 24 mm in each direction (X and Y), in 0.2 mm increments, resulting in 120 data points per direction. The alignment must be repeated for each wavelength, as the beam position shifts slightly with different wavelengths selected on the OPO. Accurate centering is crucial to obtaining reliable charge measurements and consistent results. An example of an XY scan before (Figure 4.7) and after (Figure 4.8) alignment is shown below. For later QE evaluation, we need the absolute extracted charge, calculated by subtracting the noise level from the measured charge.

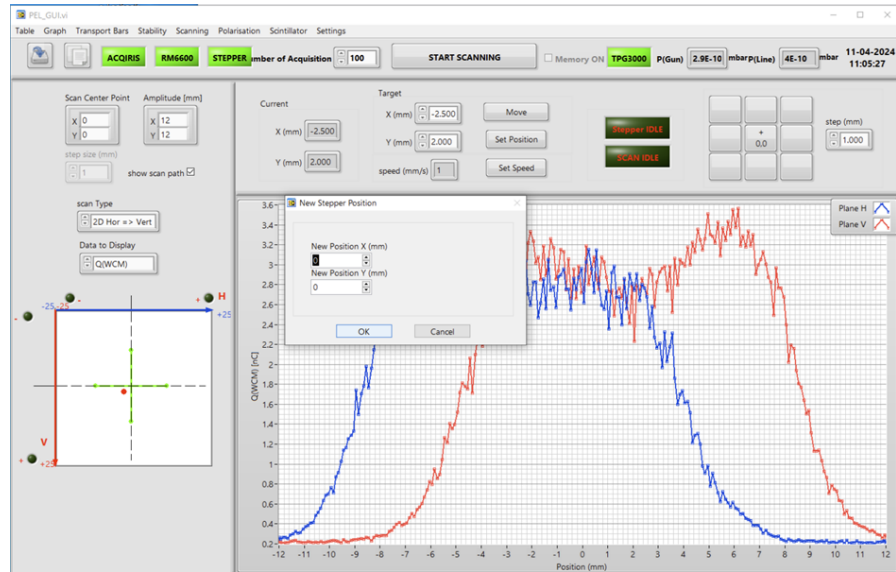


FIGURE 4.7: LabVIEW interface for centering the beam on the photocathode. Example for cathode #217 with the OPO at 266 nm, before proper alignment.



FIGURE 4.8: LabVIEW interface for centering the beam on the photocathode. Example for cathode #217 with the OPO at 266 nm, after proper alignment.

4.3 Data Acquisition & Processing

For accurate data acquisition, once the beam is centered on the cathode, 1000 laser shots are taken at each measurement point, and the extracted charge is averaged across these 1000 points. The LabVIEW interface for data acquisition displays the extracted charge based on readings from the Wall Current Monitor (WCM), Faraday Cup (FC), Fast Current Transformer (FCT) and cylinder (CYL). For later QE evaluation, only the WCM values will be used. The interface also provides real-time pressure readings in both the gun and the measurement line. Any increase in pressure may indicate laser-induced damage on the cathode surface, so it is important to monitor this parameter closely.

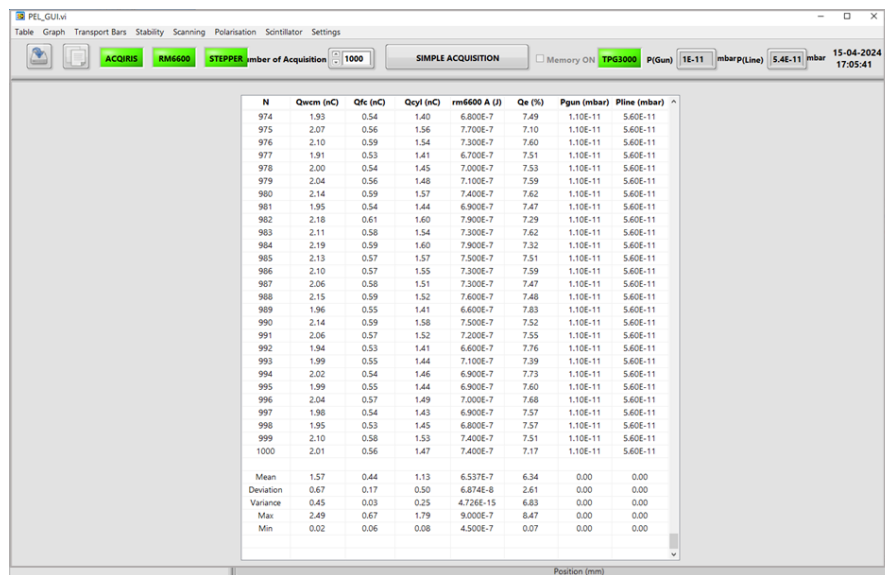


FIGURE 4.9: LabVIEW interface for data acquisition. Example for cathode #217 with the OPO at 266 nm.

Chapter 5

Results and Discussion

5.1 Results

5.1.1 QE maps

To evaluate the QE homogeneity of the rejuvenated photocathodes, surface scans were performed, scanning the laser spot in 1 mm steps over a 24 mm x 24 mm rectangular pattern in x and y direction. At each position, the extracted charge was averaged over 10 laser shots to create a detailed map of the measured quantum efficiency across the cathode surface. The results for cathode #217 are shown below. Surface scans for cathode #210 could not be performed due to time constraints.

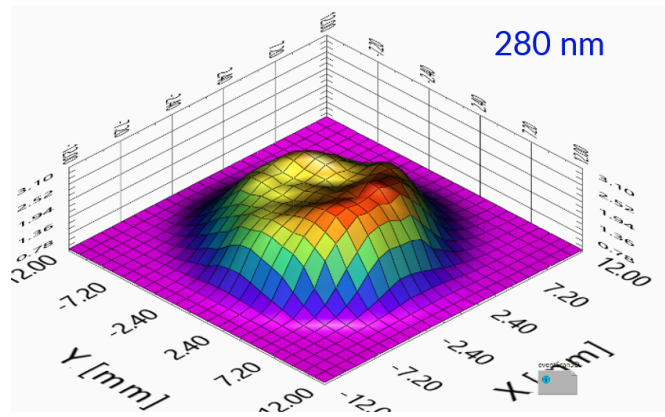


FIGURE 5.1: Surface QE map of Cathode #217 at 280 nm.

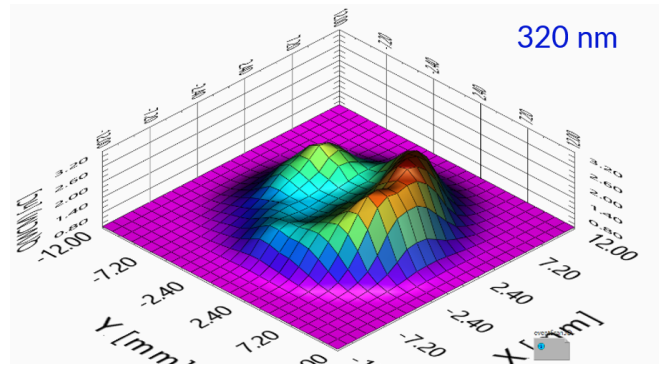


FIGURE 5.2: Surface QE map of Cathode #217 at 320 nm.

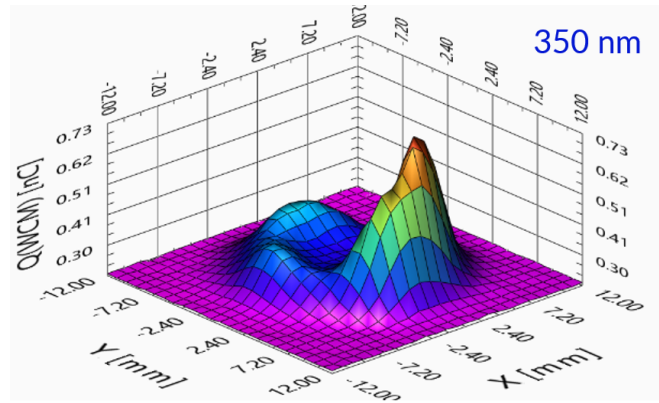


FIGURE 5.3: Surface QE map of Cathode #217 at 350 nm.

Given the considerable time required for a complete surface mapping, linear scans across the photocathode center in x and in y direction were performed after an initial full scan as a sufficient alternative to assess the amount of the extracted charge from the photocathodes at different wavelengths. The following figures show the exported data from the user interface panel corresponding to the surface scans obtained from the Wall Current Monitor measurements. The extracted charge is plotted against the position of the laser beam on the cathode, with the blue line representing the x-plane and the red line representing the y-plane.

The XY scans for both cathodes at selected wavelengths (240, 300, 350, and 400 nm) are presented in this chapter to provide an overview of the observed trends. Figures 5.4-5.7 present the scans for cathode #217, while Figures 5.8-5.11 show the data for cathode #210. Additional scans covering a broader wavelength range from 250 to 430 nm are provided in Appendix C for reference.

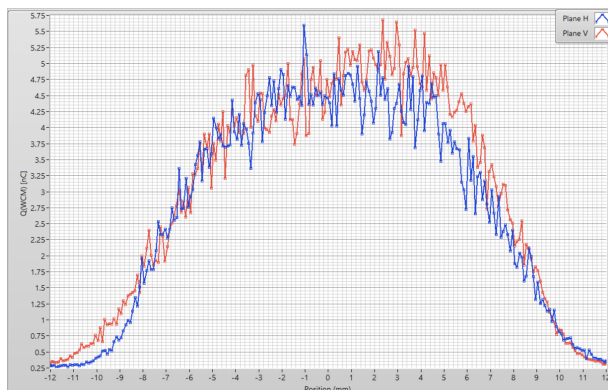


FIGURE 5.4: Surface QE map of Cathode #217 at 240 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $0.66 \mu\text{J}$.)

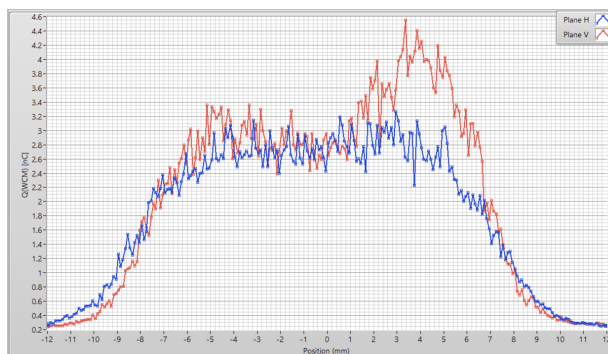


FIGURE 5.5: Surface QE map of Cathode #217 at 300 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $0.92 \mu\text{J}$.)

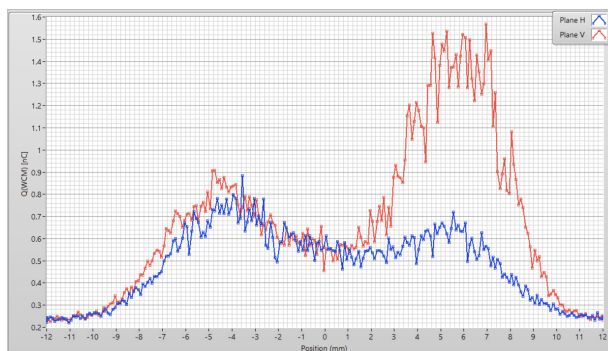


FIGURE 5.6: Surface QE map of Cathode #217 at 350 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $14.4 \mu\text{J}$.)

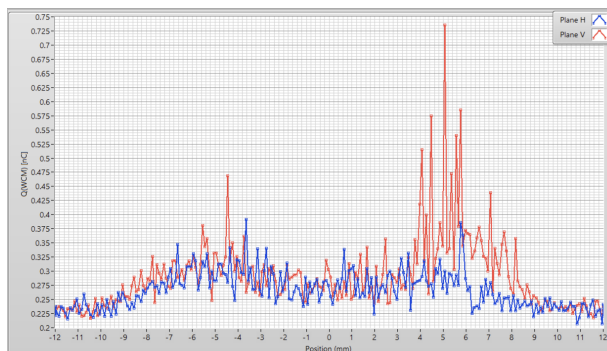


FIGURE 5.7: Surface QE map of Cathode #217 at 400 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $58.0 \mu\text{J}$.)

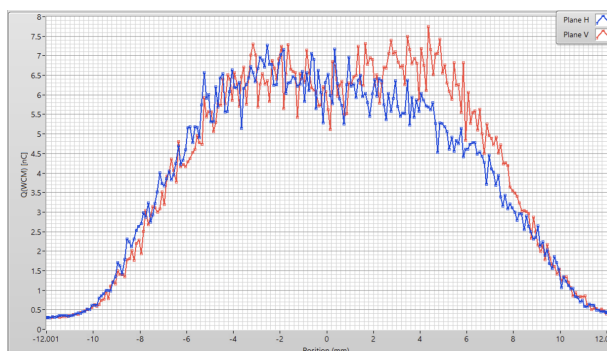


FIGURE 5.8: Surface QE map of Cathode #210 at 240 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $0.64 \mu\text{J}$.)

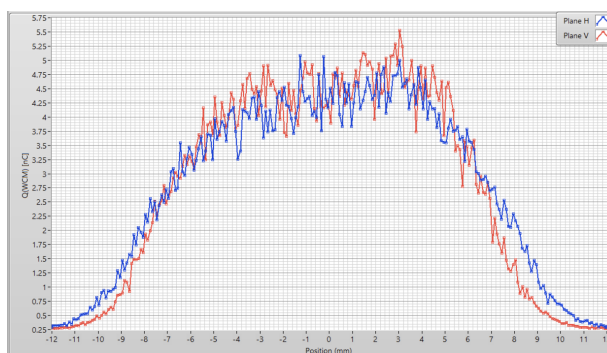


FIGURE 5.9: Surface QE map of Cathode #210 at 300 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $0.60 \mu\text{J}$.)

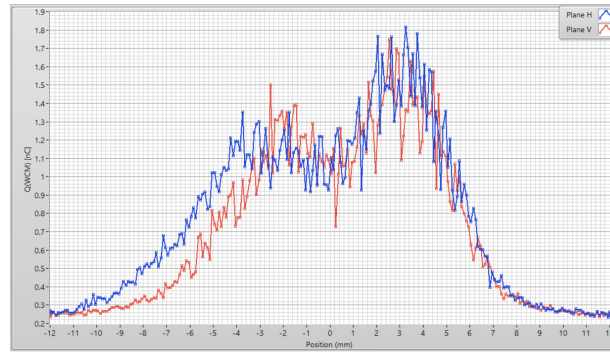


FIGURE 5.10: Surface QE map of Cathode #210 at 350 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $2.21 \mu\text{J}$.)

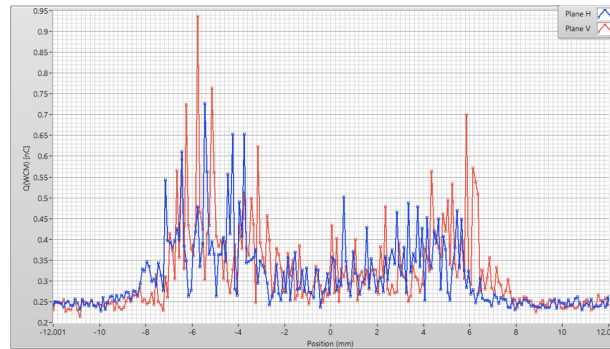


FIGURE 5.11: Surface QE map of Cathode #210 at 400 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $3.60 \mu\text{J}$.)

5.1.2 Spectral Response Data

To accurately determine the quantum efficiency at each wavelength, the quantum yields were measured at six different attenuation factors corresponding to varying laser energies. Data points were recorded for each configuration and the values of the non-saturated leftmost region, showing a linear trend were selected to calculate the QE at the given wavelength using Equation 2.2.

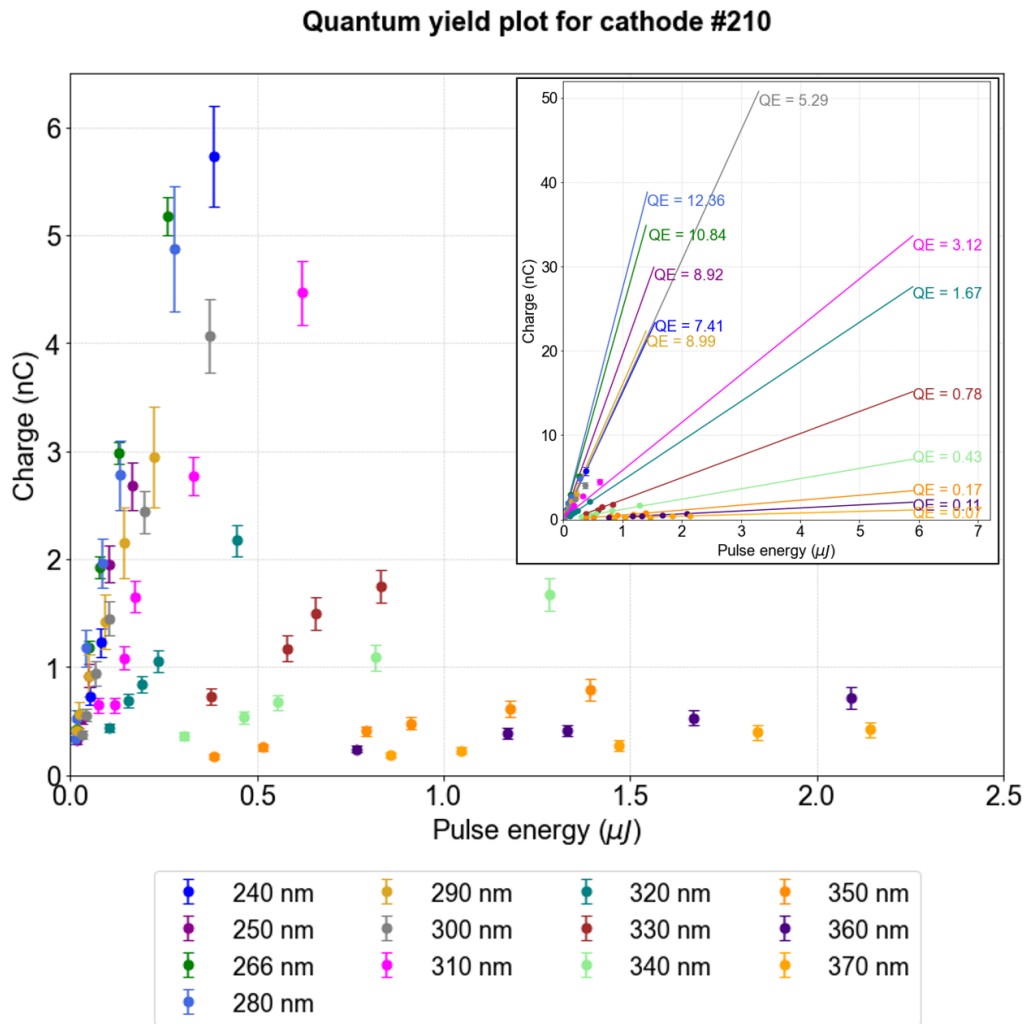


FIGURE 5.12: Quantum yield plot for Cathode #210

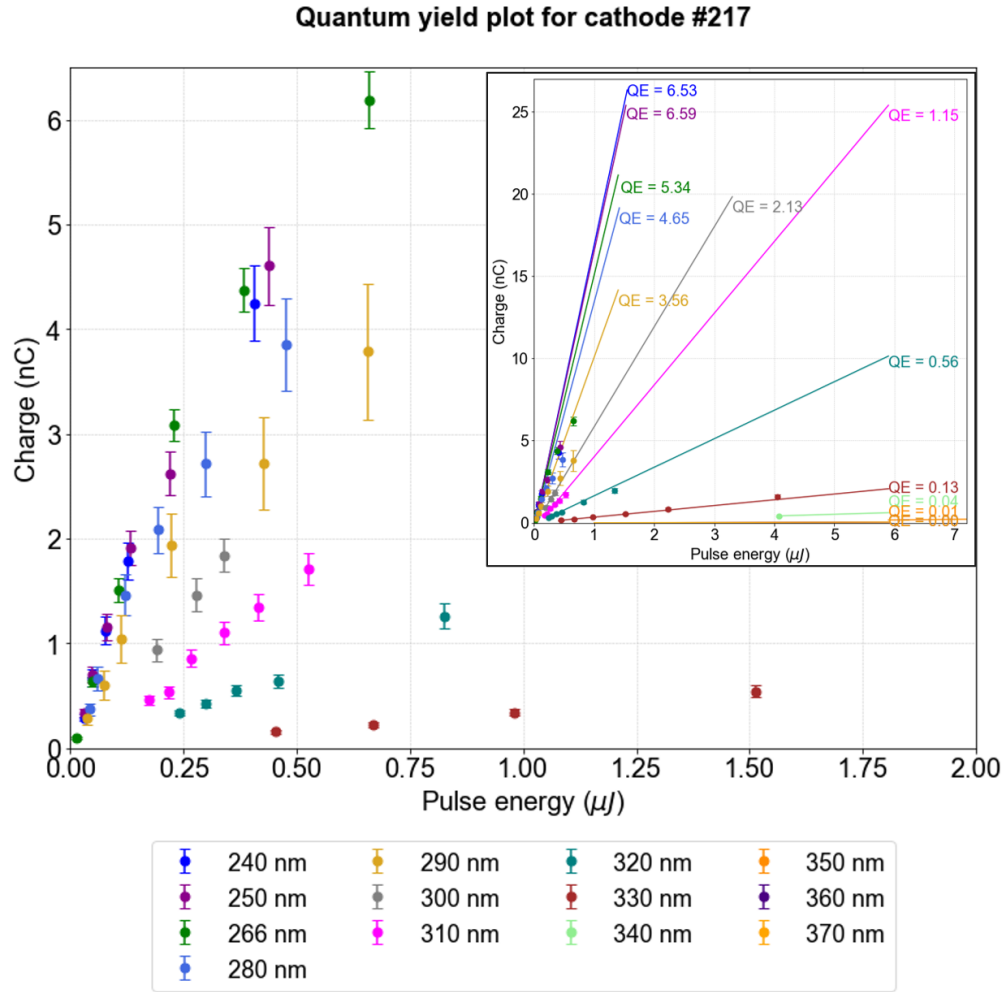


FIGURE 5.13: Quantum yield plot for Cathode #217

The QE values calculated for each wavelength were plotted against the corresponding photon energies to analyze the spectral response of the cathodes. Figures 5.14 and 5.15 show these results, both on a normal scale and on a logarithmic scale. For a complete list of QE values, see Appendix D.

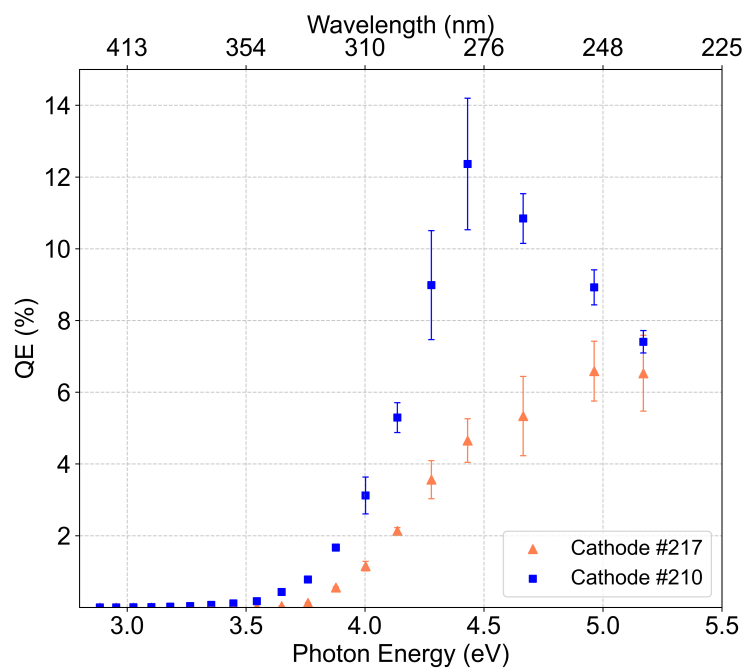
Spectral response of cathodes #210 and #217

FIGURE 5.14: Spectral response values of Cathode #210 and #217.

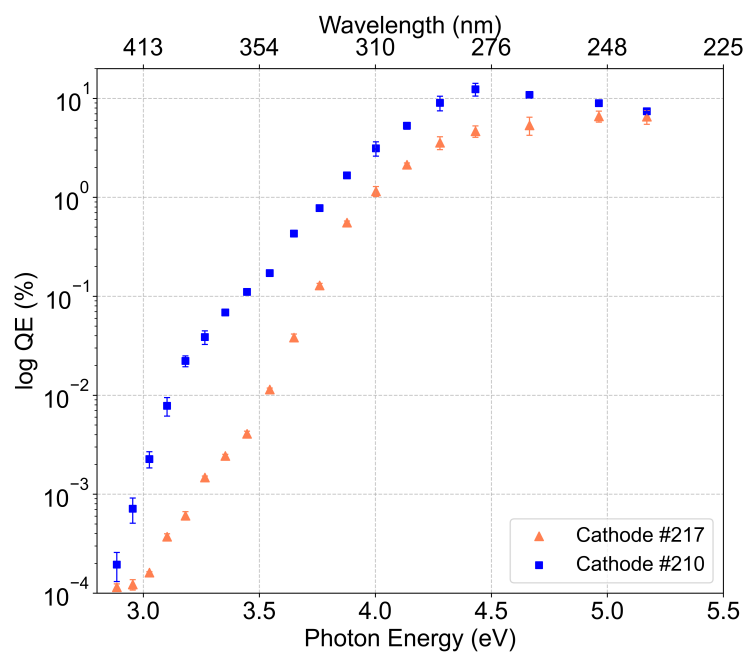
Spectral response of cathodes #210 and #217

FIGURE 5.15: Spectral response values of Cathode #210 and #217 (logarithmic scale).

5.1.3 Fitting of the Three-Step Model

To evaluate how well the results agree with Spicer's Three-Step Model for semiconductors, we applied the equation 2.9 to the data points. For convenience, the equation is restated below:

$$QE(\hbar\omega) \approx \frac{B}{g + \frac{g}{(\hbar\omega - V_g)^v}},$$

where $QE(\hbar\omega)$ is the quantum efficiency as a function of photon energy ($\hbar\omega$), B and g are fitting parameters, V_g is the threshold energy, and v is determined by Spicer's model to be equal to $v = \frac{2}{3}$ [35]. The fitting was applied separately to both the high-energy and low-energy data points, as these correspond to two distinct photoemissive phases. The Three-Step Model was initially fitted using all available data points and subsequently refined by removing outlier data points. This methodology of selectively omitting certain data points to achieve a more accurate fit was based on the approach described in the work of Chevallay et al. [75] and in the dissertation work of Trautner [48].

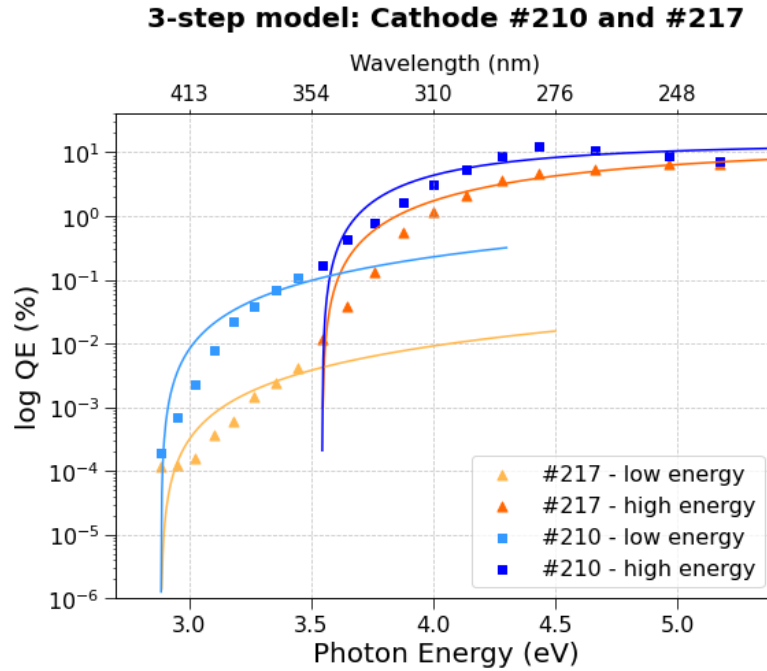


FIGURE 5.16: Fitting Spicer's 3-step model to the spectral response curves for cathodes #210 and #217, using all data points.

TABLE 5.1: Fitting parameters for Spicer's Three-Step Model applied to Cathodes #210 and #217, using all available data points.

Energy range	Cathode	B	g	V_g
Low energy	#210	3.720	6.864	2.886
	#217	0.421	14.375	2.886
High energy	#210	15.020	0.829	3.545
	#217	18.071	2.134	3.545

In Figure 5.17, the lowest energy data points were omitted from each fitting curve to assess whether reducing outlier points improved the fit to the data.

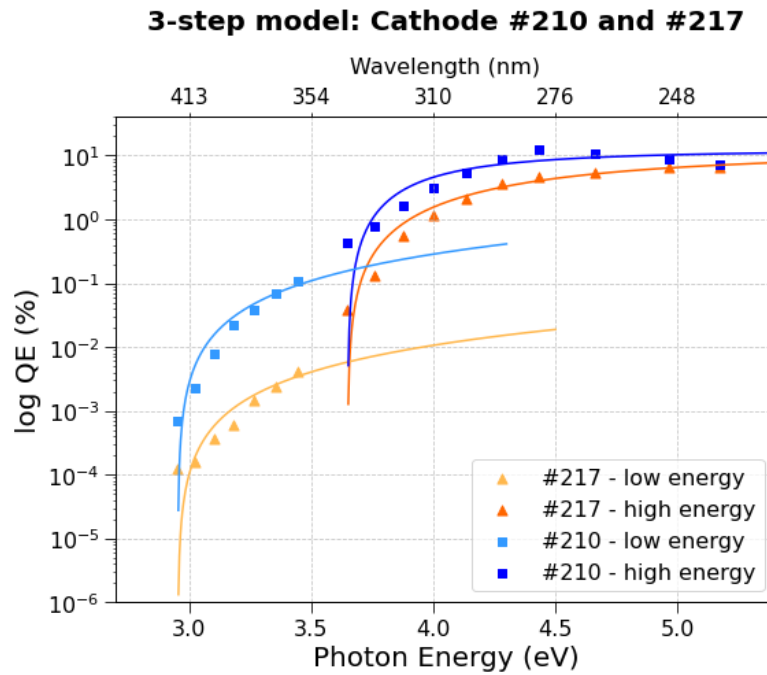


FIGURE 5.17: Fitting Spicer's 3-step model to the spectral response curves for cathodes #210 and #217, omitting the lowest energy data point for each fitting curve.

TABLE 5.2: Fitting parameters for Spicer's Three-Step Model applied to Cathodes #210 and #217, omitting the lowest energy data point for each fitting curve.

Energy range	Cathode	B	g	V_g
Low energy	#210	12.652	12.917	2.954
	#217	0.563	14.563	2.954
High energy	#210	12.748	0.518	3.649
	#217	12.733	1.311	3.649

For comparison, the plots of similar curves using the Three-Step Model approximation from the works of Chevallay et al. [75] and Trautner [48] are presented below. Data points were digitally extracted from the original publications and replotted. The corresponding curves according to the three-step model were generated to approximate the original curves. Since the exact fitting parameters were not provided in the original papers, the parameters presented in Table 5.3 may differ; however, the approximation effectively reproduces the original data trends.

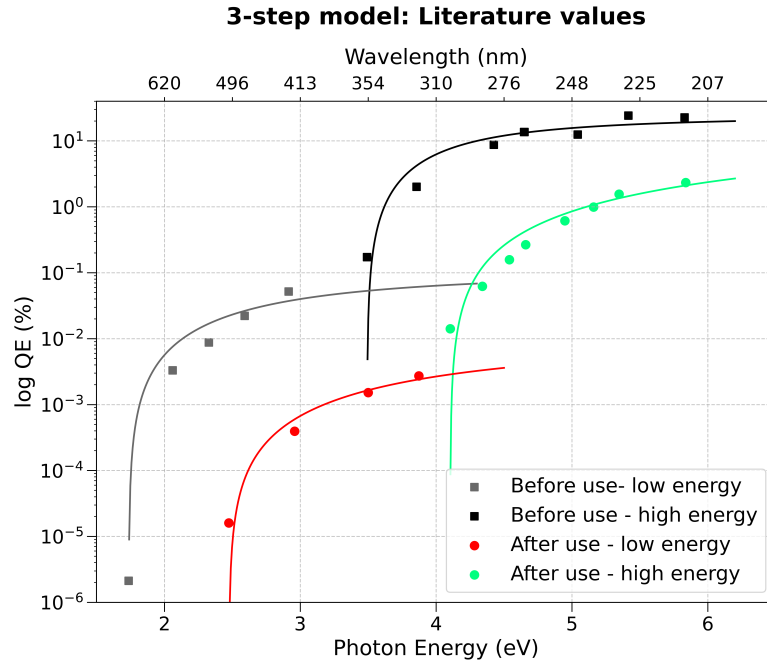


FIGURE 5.18: Fitting of Spicer's three-step model to the spectral response values for Cs-Te cathodes before and after use in the RF gun. Data reported by Chevallay et al. [75].

TABLE 5.3: Fitting parameters used to approximate the photoemission threshold in the Three-Step Model for the values reported by Chevallay et al. [75].

Energy range	Cathode	B	g	V_g
Low energy	Before use	0.110	1.847	1.737
	After use	0.011	3.209	2.474
High energy	Before use	24.756	1.051	3.492
	After use	16.290	6.183	4.104

As the next step, the Three-Step Model curves from this investigation were plotted along with the literature data for direct comparison. For our analysis, the fitting curves generated using reduced number of data points were used as they provided a better fit to the observed trends. This approach allows a qualitative description of the photocathodes and a comparison with literature data.

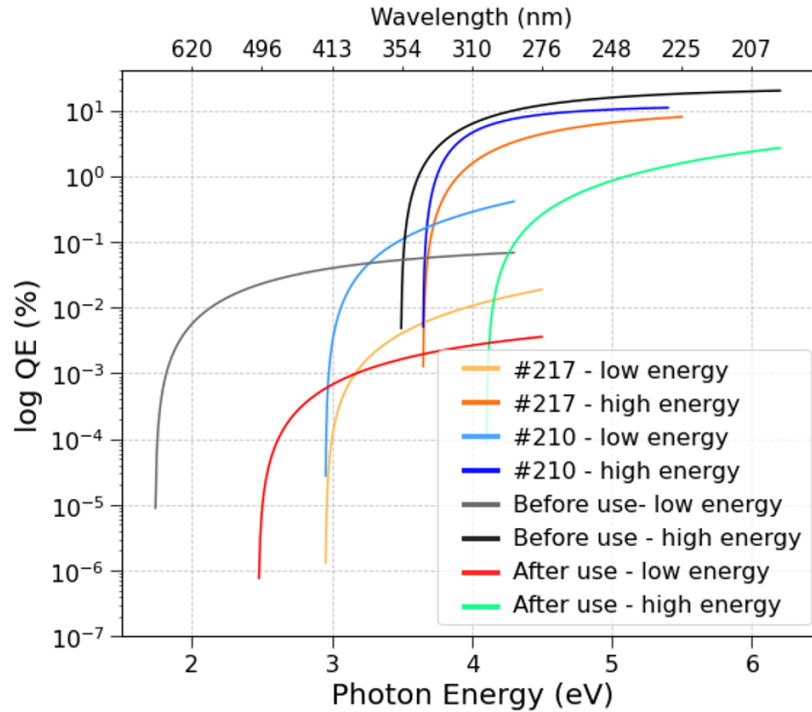


FIGURE 5.19: Comparison of the Three-Step Model approximation for Cs-Te photocathodes determined in this investigation with values reported in the studies by Chevallay et al. [75] and Trautner [48].

5.2 Discussion

5.2.1 QE maps

The XY scans for both rejuvenated photocathodes (Cathode #210 and #217) show charge profiles that define the regions where the cathodes are coated with the photoemissive thin film compounds. This coated region has a diameter of approximately 20 mm for both cathodes, with a transition region at the edges, where the charge gradually increases. For both cathodes, the highest extracted charge is not located in the center, but rather along the edges near the central region.

For Cathode #217, the surface scans at 240 nm (see Figure 5.4) show a relatively flat charge distribution across the cathode surface, as observed in both the XY scan and the 3D map. However, at higher wavelengths the charge distribution becomes increasingly inhomogeneous and a gradual trend emerges, with the edges in both the X and Y directions showing increased charge extraction at lower photon energies. In addition, a particularly pronounced enhancement is observed in the Y direction between 2 and 8 cm, which becomes noticeable above 250 nm and highly pronounced at 300 nm and above (see Figures 5.5 - 5.7). This strong inhomogeneity is further highlighted in the QE maps at 320 nm and 350 nm, where a distinct region of increased charge is consistently observed at the same location (see Figures 5.2 and 5.3).

In the case of cathode #210, the XY scans show an overall good homogeneity of the charge distribution at 240 nm and 250 nm (see Figure 5.8). However, at higher wavelengths, a region along the edge between 2 and 6 cm becomes more pronounced in both X and Y directions on both sides. Between 380 nm and 430 nm, this inhomogeneous region becomes more balanced and shows less enhancement of the edges (see Figures 5.10 - 5.11). At 430 nm, the extracted charge approaches the noise level, making it difficult to center the cathode in the XY plane.

Regions where more charge can be extracted at lower photon energies may be attributed to localized excess deposition of cesium. This excess lowers the work function, allowing the emission of low energy electrons. Such uneven distribution is likely influenced by deposition parameters, such as the positioning of dispensers during fabrication. However, since the scan provides only a qualitative measure, the observed enhancements cannot be attributed solely to uneven Cs-Te deposition, leaving the exact

origin of these features uncertain. To verify that these enhancements are indeed caused by overcesiation, a detailed surface analysis, for example using XPS, would be required to better understand the material composition and its distribution over the cathode surface.

For both cathodes, the central region of the rejuvenated thin film coating shows a dip in charge extraction compared to the edges. This feature could be indicative of laser-induced damage, electron back-bombardment, or other mechanisms of thin film degradation due to charge extraction. However, the overall recovery of quantum efficiency values above 10 % indicates that rejuvenation via co-deposition of used and pre-RF conditioned photocathodes without breaking the UHV conditions is a feasible option.

5.2.2 Spectral Response Data

The spectral response curves for cathodes #210 and #217 exhibit similar characteristics, including two distinct shoulders and a declining edge at lower photon energies (see Figure 5.15). The high-energy shoulder is observed above 3.5 eV, while the low-energy shoulder occurs below 3.5 eV. This characteristic is consistent with reports in the literature where overcesiated Cs-Te cathodes exhibit similar behavior. These shoulders are typically attributed to the coexistence of two separate Cs-Te phases: Cs_2Te , which is dominant in the high-energy range, and another phase, such as Cs_5Te_3 or metallic Cs, which contributes to the low-energy shoulder.

In the present study, the stoichiometric deposition ratios during the first fabrication of both cathodes were approximately 4:1 (see Table 3.7), significantly exceeding the ideal ratio of 2:1 for Cs_2Te . This over-deposition suggests that there is an excess of cesium on the surface, since not all of the cesium can fully react to Cs_2Te . After rejuvenation with a new Cs-Te layer, the stoichiometric deposition ratios were reduced to 1.24 for cathode #210 and to 3.27 for cathode #217. The lower ratio for cathode #210 implies less residual cesium compared to cathode #217. However, despite this difference, cathode #210 still shows a pronounced low-energy shoulder. This observation could be explained by contributions from the original Cs-Te layer beneath the rejuvenated film, since cathode #210 was not exposed to other chemical contamination after its initial production, which could reduce the emission threshold. These results suggest that the rejuvenated cathodes may retain some characteristics of their original layers,

which could affect their spectral response. To further investigate this behavior, surface analysis by XPS and complementary argon ion beam depth profiling could be beneficial to quantify the presence of excess caesium in the thin film layer.

A comparison of the two cathodes reveals significant differences in their maximum QE values and corresponding wavelengths. Cathode #210 achieves a peak QE of 12.4% at 280 nm, while cathode #217 reaches a peak QE of 6.59% at 250 nm. Across the spectrum, cathode #210 consistently exhibits higher QE values than cathode #217. This can be attributed to differences in their manufacturing, storage, and rejuvenation conditions, including variations in stoichiometric deposition ratios and the oxygen exposure experienced by cathode #217 during a vacuum degradation incident in the RF-gun. While these factors likely contributed to the degradation of the QE of cathode #217, the exact mechanisms responsible for its lower performance remain unclear due to the complexity of the influencing variables. In addition, the consistently higher spectral response of cathode #210 may reflect a more uniform distribution of Cs-Te phases or greater resistance to contamination and degradation over time.

5.2.3 Fitting of the Three-Step Model

The purpose of fitting the Three-Step Model is to determine the threshold energy of the material and compare it with literature values to identify the compounds present on the cathode surface. Initial attempts to apply the model to all data points, both in the high and in the low energy regions, did not provide a satisfactory fit. This discrepancy is likely due to the cumulative spectral response of multiple materials on the cathode surface. In order to better characterise the dominant materials contributing to photoemission, the model was refined by excluding the lowest energy data points, resulting in a significantly improved fit. The data point at 430 nm was omitted for the low-energy fitting, and the 350 nm point was excluded for the high-energy fitting.

With the Three-Step Model, the parameter V_g - which characterizes the photoemission threshold of the material - was determined to be 2.9 eV for the low-energy shoulder and 3.6 eV for the high-energy shoulder. However, the fitting parameters, including V_g , were found to be highly dependent on the fitting procedure and the data points included. The lowest energy data point used in the fit plays a significant role in determining the threshold energy. For this reason, the results of the Three-Step Model fitting must be critically evaluated, taking into account the influence of the selected data set.

In the literature, V_g is described as the threshold energy of the material below which no photoemission occurs. However, in the case of a photocathode material exhibiting two distinct shoulders - indicating two photoemissive phases and requiring two fitting curves - the determination of two V_g parameters as threshold energies is contradictory, since in this case electron emission occurs below the V_g value of the high-energy shoulder. For this reason, we will refer to V_g as a parameter to describe the photoemissive phases present on the photocathodes, according to the Three-Step Model.

For the high-energy shoulder, V_g values of 3.6 eV were obtained for both cathode #210 and cathode #217. These values are in close agreement with literature data where a V_g value of 3.5 eV is attributed to the compound Cs_2Te in caesium-rich photocathodes, which typically exhibit two distinct photoemission shoulders [59]. This consistency suggests that the material responsible for the high-energy shoulder is likely Cs_2Te .

In contrast, for the low energy shoulder, V_g values of 2.9 eV were determined for both cathodes, which are significantly higher than the 1.7 eV reported in the literature for fresh cathodes and the 2.5 eV for used cathodes. In the work of Chevallay et al. [75] and Trautner [48], the shift in V_g for used cathodes was attributed to laser-induced damage or electric field effects during operation, and their results showed that the entire spectral response curve shifted to higher photon energies, not just the low-energy shoulder. However, in this investigation, the high energy V_g values for cathode #210 and cathode #217 (3.6 eV) are consistent with literature values for fresh cathodes. This indicates that the high energy shoulder remains unaffected and that the increase in V_g is only found in the low energy shoulder.

The higher V_g values observed for the low-energy shoulder in this study are likely due to experimental limitations rather than intrinsic properties of the photocathodes. In particular, V_g is highly dependent on the lowest energy data point included in the fit. In this study, no data could be recorded at photon energies below 3.0 eV because the extracted charge was too low to be measured with sufficient accuracy.

Chapter 6

Conclusions and Outlook

6.1 Conclusion

This thesis presents a qualitative investigation of Cs-Te photocathodes, with a particular focus on their spectral response in relation to their material composition. A systematic study was conducted on rejuvenated Cs-Te photocathodes obtained via co-deposition as a rejuvenation method. This approach successfully restored the quantum efficiency of degraded photocathodes to a significant extent, demonstrating its applicability as a quick film replacement technique without breaking the UHV conditions, especially when performed in situ.

The spectral response analysis revealed two distinct features in the response curves of the photocathodes, corresponding to two different photoemissive phases. Spicer's three-step model effectively described the obtained high-energy data of the rejuvenated Cs-Te photocathodes, confirming the presence of the stoichiometric compound of Cs_2Te on the cathode surface. However, the model had limitations in approximating the photoemission threshold at lower energies, likely due to experimental constraints. These limitations could be addressed by improving the sensitivity of the laser energy detection, increasing the laser spot size and improving the laser beam profile. Since the emission threshold for the low-energy shoulder could not be precisely determined and no further surface or depth profiling analysis was carried out, its origin remains speculative. A possible explanation is the presence of excess cesium, which could alter the photocathode's electronic properties. Excess cesium at the $\text{Cs}_2\text{Te}/\text{Cs}$ interface

appears to lower the material's threshold barrier, thus enhancing electron emission in the low energy range. This behavior is consistent with cesium's Negative Electron Affinity (NEA) effects observed in other semiconductor photocathodes [47]. Such NEA effects likely contribute to the observed low-energy shoulder, as they influence the energy barrier for photoemission and shift the emission threshold.

To gain a deeper understanding of the low-energy spectral features, further investigations are needed. These should include extending the measurements to lower photon energies and performing surface-sensitive analyses such as XPS. Such improvements would provide direct evidence to confirm the origin of the low-energy shoulder. If this feature is confirmed to be due to excess cesium, it would allow the development of a more precise Cs-Te deposition process. Real-time monitoring using multi-wavelength low-energy laser diagnostics in the range around the material emission threshold during production could detect charge emission at photon energies associated with the low-energy shoulder. This would allow precise control and shutdown of the cesium dispenser when a charge in this range is detected, thus identifying excess deposition. This approach would optimize the control of the stoichiometric deposition ratio. By observing and possibly eliminating the formation of the low-energy shoulder, this method could improve the chemical composition of the Cs-Te thin film layer. Such an optimized layer composition would reduce the Mean Transverse Energy (MTE) of the extracted electron bunches and increase beam brightness, thereby improving the quality of the electron source.

To further investigate photocathodes and refine their characterization, the planned installation of the transverse energy spread spectrometer (TESS) [76] system in the photoemission laboratory will provide a valuable addition to the measurement capabilities of the lab. This diagnostic tool is designed to measure the MTE of electron beams with high precision. Monitoring the MTE will provide deeper insight into beam dynamics, enabling improved characterization techniques and guiding the production of higher quality photocathodes.

This study investigates the spectral response of photocathodes rejuvenated by the co-deposition method. The lack of comparative data for photocathodes rejuvenated by this approach underscores the need for further research. Future work should focus on quantitative characterisation of the photocathode coating to correlate with the spectral response data to provide a clearer understanding of the components present on the cathode surface.

Appendix A

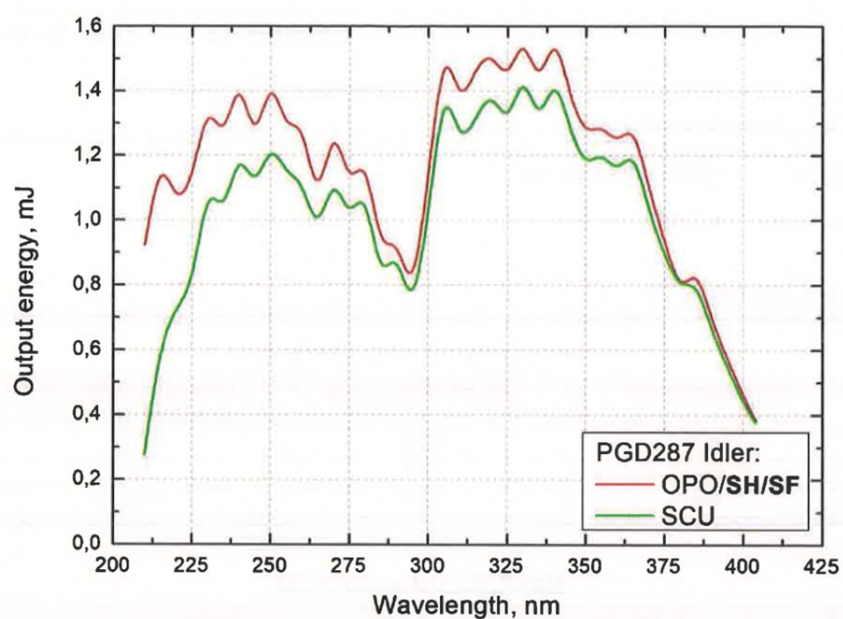


FIGURE 1: SH/SF output energy as a function of wavelength (diagram adapted from the EKSPLA manual).

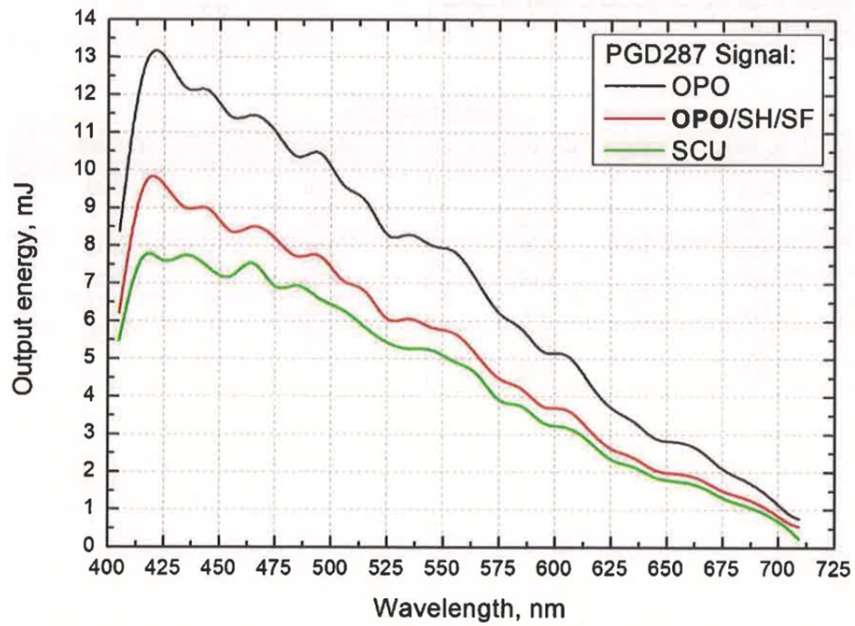


FIGURE 2: Signal output energy as a function of wavelength (diagram adapted from the EKSPLA manual).

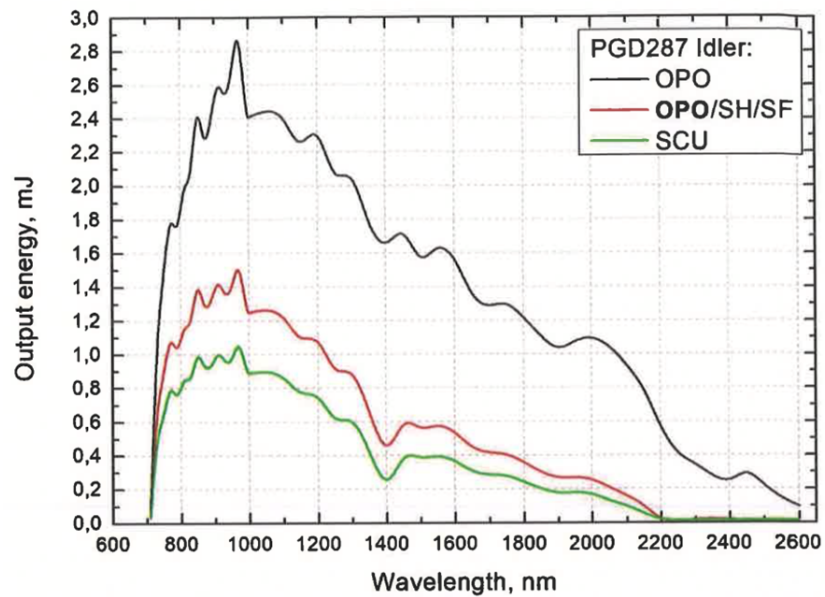


FIGURE 3: Idler output energy as a function of wavelength (diagram adapted from the EKSPLA manual).

Appendix B

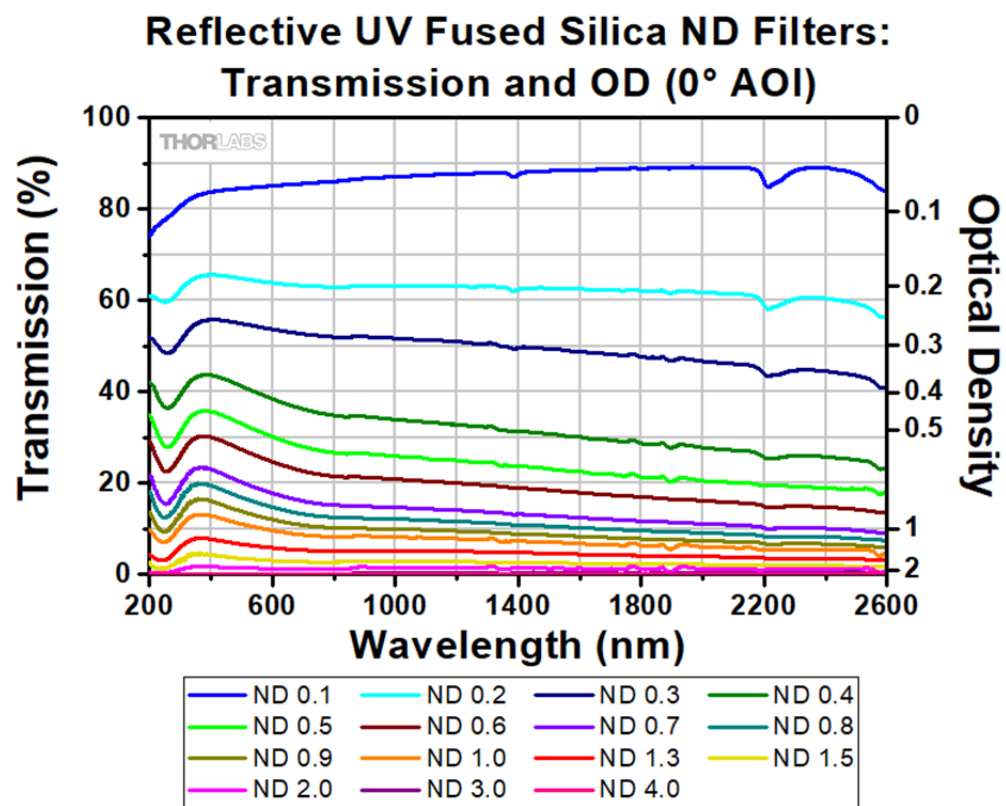


FIGURE 4: Transmission and Optical Density Characteristics of Reflective UV Fused Silica Neutral Density (ND) Filters from ThorLabs [73]

Appendix C

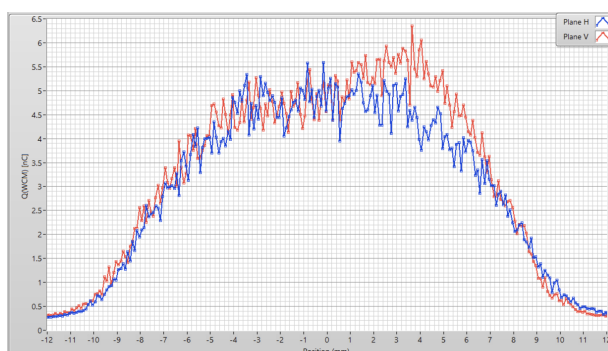


FIGURE 5: Surface QE map of Cathode #217 at 250 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $0.71 \mu\text{J}$.)

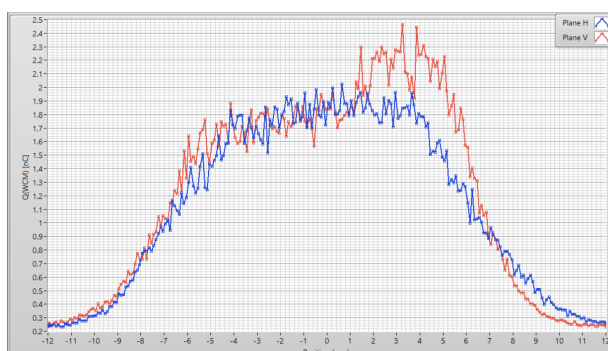


FIGURE 6: Surface QE map of Cathode #217 at 266 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $0.17 \mu\text{J}$.)

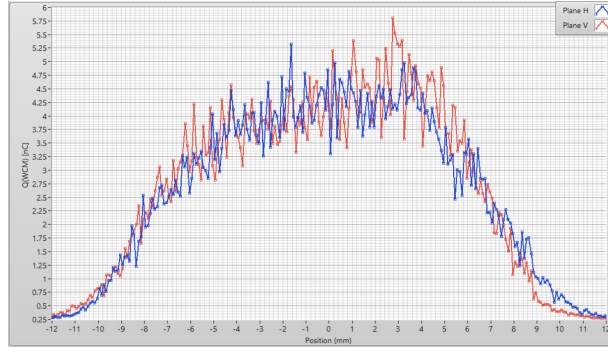


FIGURE 7: Surface QE map of Cathode #217 at 280 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $0.78 \mu\text{J}$.)

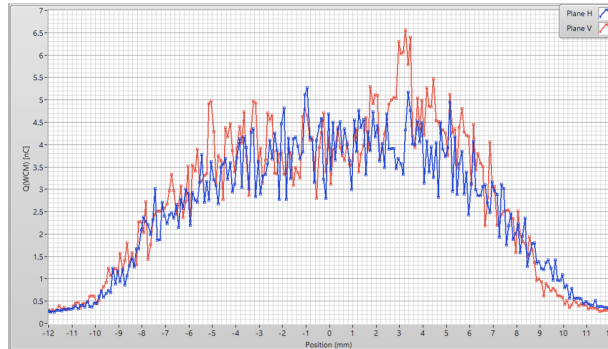


FIGURE 8: Surface QE map of Cathode #217 at 290 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $1.06 \mu\text{J}$.)

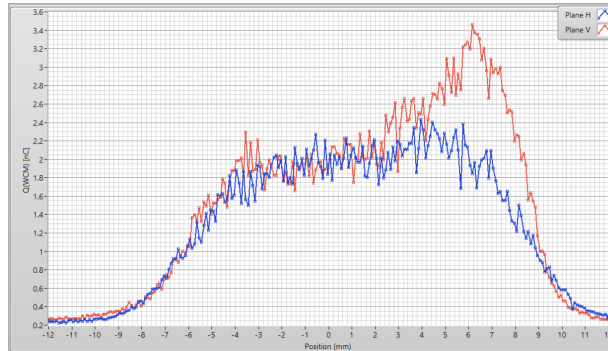


FIGURE 9: Surface QE map of Cathode #217 at 310 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $0.85 \mu\text{J}$.)

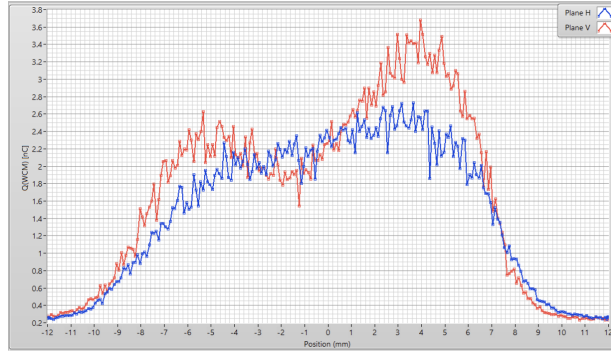


FIGURE 10: Surface QE map of Cathode #217 at 320 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $2.16 \mu\text{J}$.)

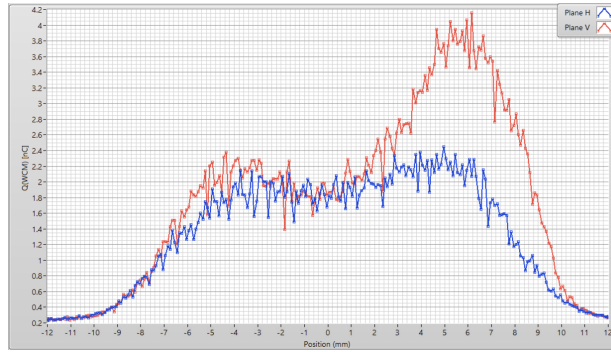


FIGURE 11: Surface QE map of Cathode #217 at 330 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $6.54 \mu\text{J}$.)

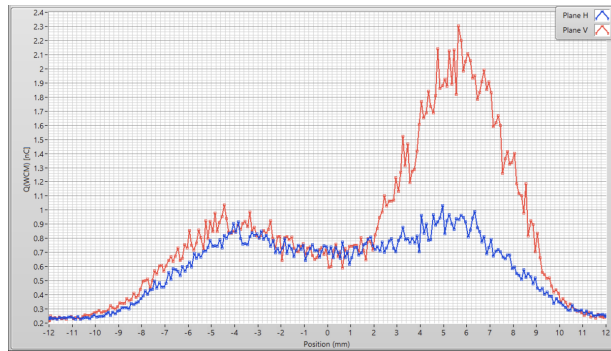


FIGURE 12: Surface QE map of Cathode #217 at 340 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $6.8 \mu\text{J}$.)

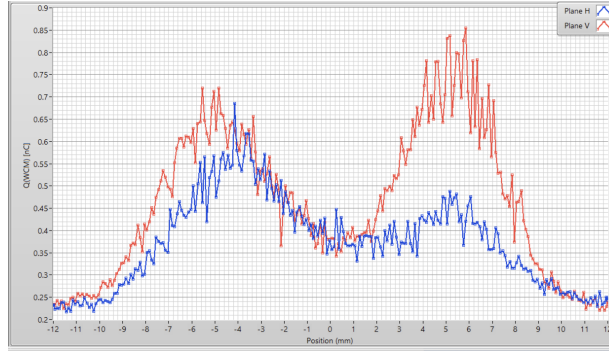


FIGURE 13: Surface QE map of Cathode #217 at 360 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $53.0 \mu\text{J}$.)

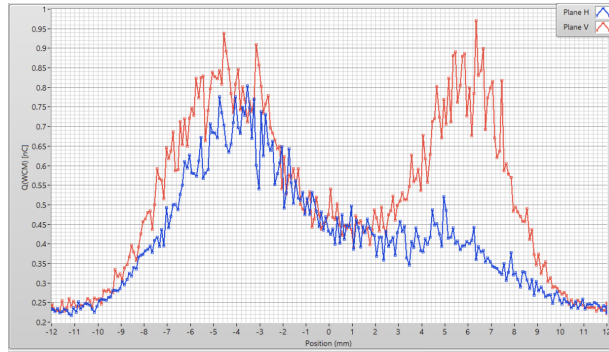


FIGURE 14: Surface QE map of Cathode #217 at 370 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $46.9 \mu\text{J}$.)

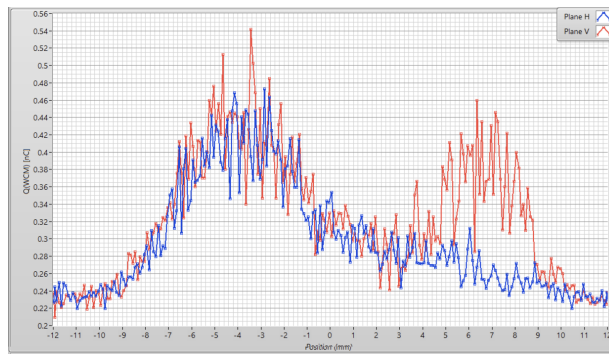


FIGURE 15: Surface QE map of Cathode #217 at 380 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $29.8 \mu\text{J}$.)

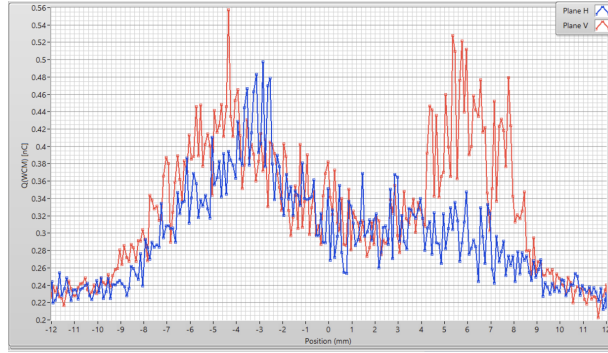


FIGURE 16: Surface QE map of Cathode #217 at 390 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $55.0 \mu\text{J}$.)

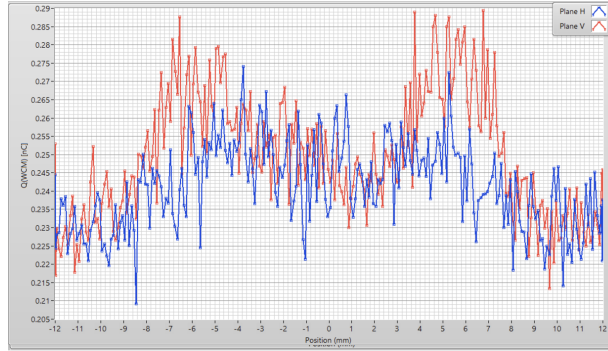


FIGURE 17: Surface QE map of Cathode #217 at 410 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $57.3 \mu\text{J}$.)

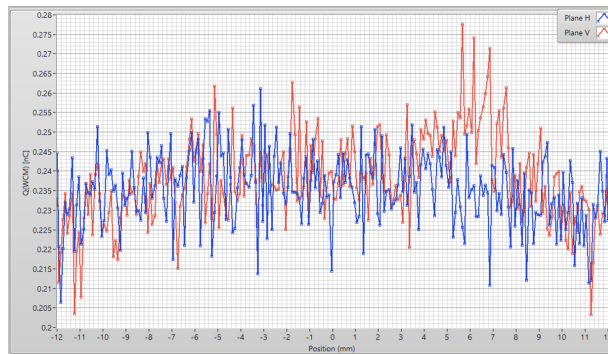


FIGURE 18: Surface QE map of Cathode #217 at 420 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $87.0 \mu\text{J}$.)

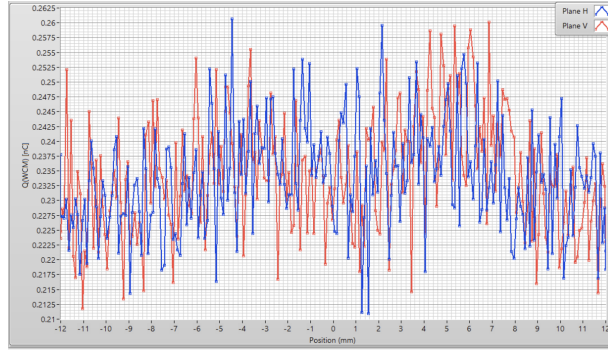


FIGURE 19: Surface QE map of Cathode #217 at 430 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $74.6 \mu\text{J}$.)

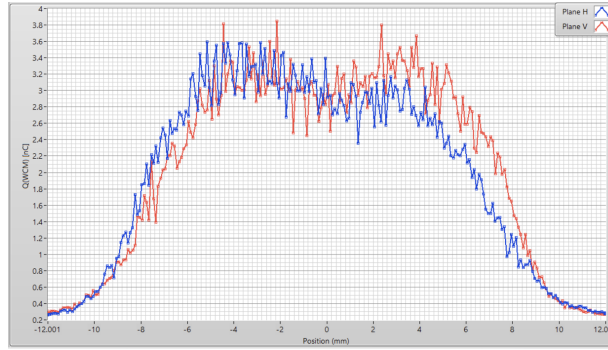


FIGURE 20: Surface QE map of Cathode #210 at 250 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $0.27 \mu\text{J}$.)

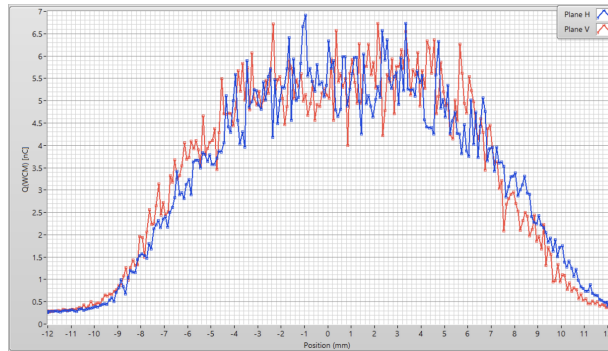


FIGURE 21: Surface QE map of Cathode #210 at 280 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $0.46 \mu\text{J}$.)

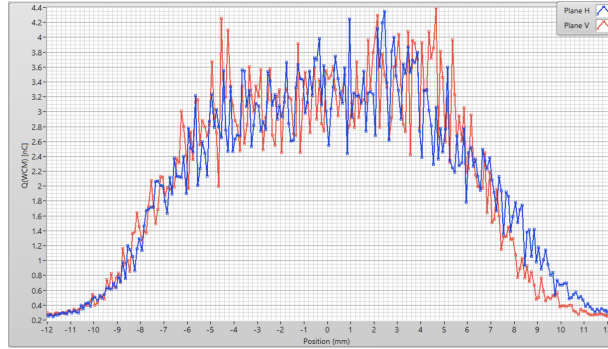


FIGURE 22: Surface QE map of Cathode #210 at 290 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $0.36 \mu\text{J}$.)

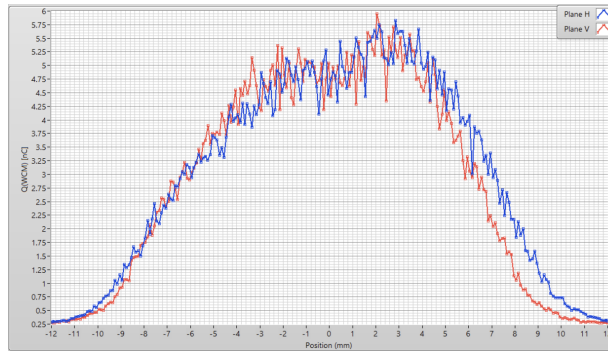


FIGURE 23: Surface QE map of Cathode #210 at 310 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $1.00 \mu\text{J}$.)

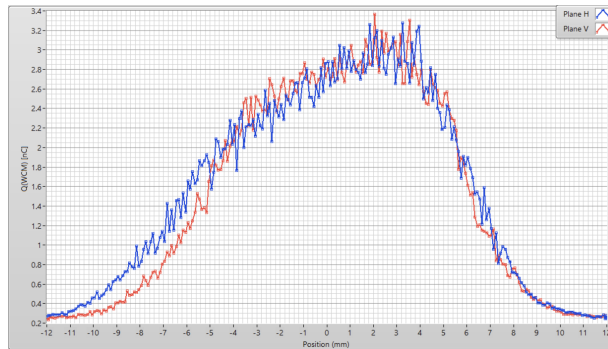


FIGURE 24: Surface QE map of Cathode #210 at 320 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $0.72 \mu\text{J}$.)

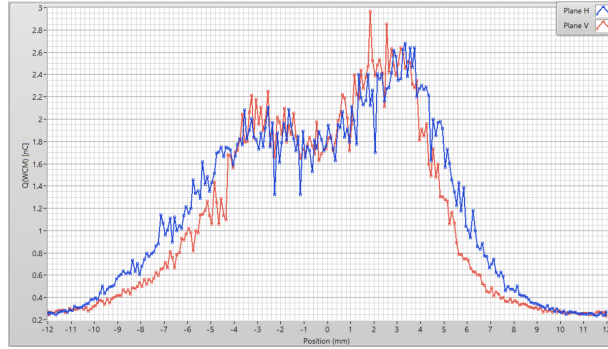


FIGURE 25: Surface QE map of Cathode #210 at 330 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $1.07 \mu\text{J}$.)

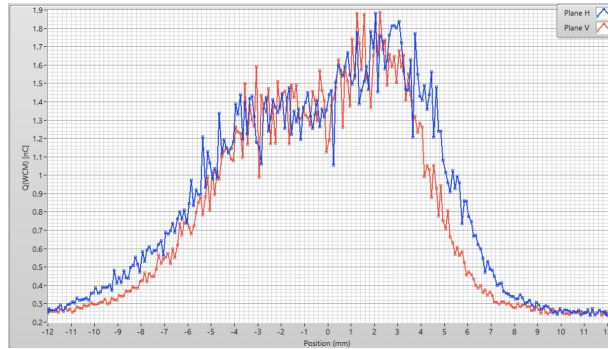


FIGURE 26: Surface QE map of Cathode #210 at 340 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $1.30 \mu\text{J}$.)

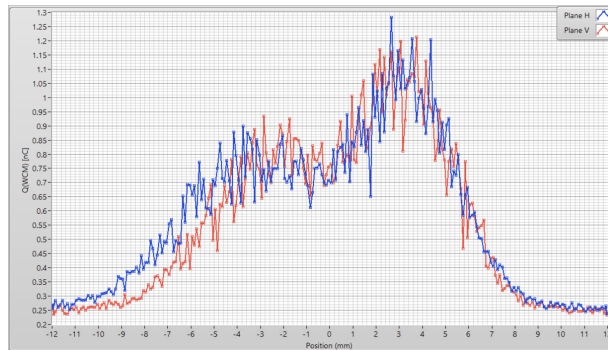


FIGURE 27: Surface QE map of Cathode #210 at 360 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $2.65 \mu\text{J}$.)

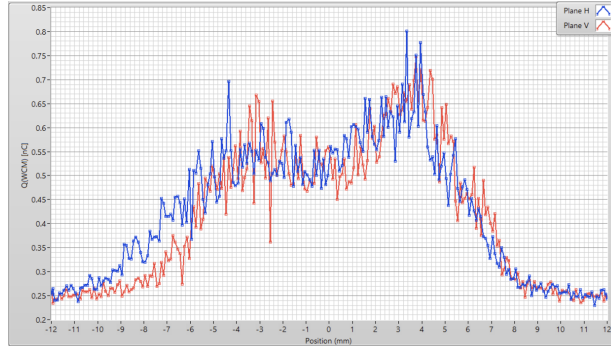


FIGURE 28: Surface QE map of Cathode #210 at 370 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $2.38 \mu\text{J}$.)

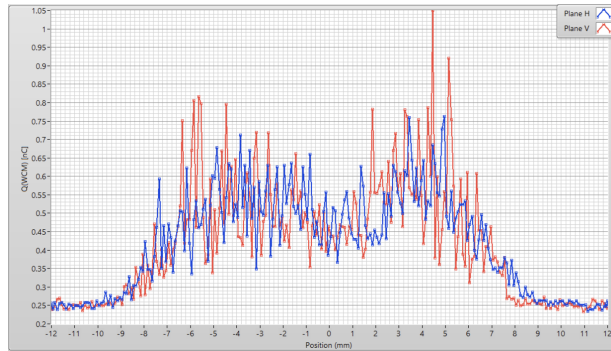


FIGURE 29: Surface QE map of Cathode #210 at 380 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $2.90 \mu\text{J}$.)

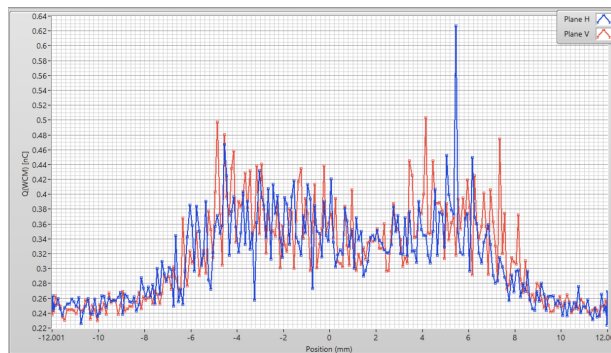


FIGURE 30: Surface QE map of Cathode #210 at 390 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $2.30 \mu\text{J}$.)

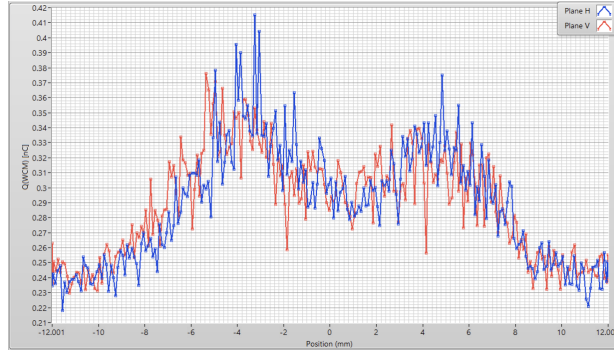


FIGURE 31: Surface QE map of Cathode #210 at 410 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $8.20 \mu\text{J}$.)

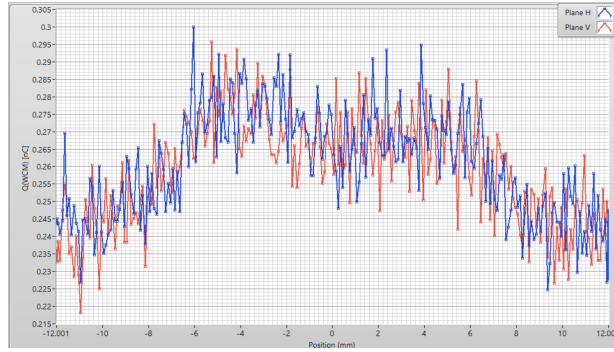


FIGURE 32: Surface QE map of Cathode #210 at 420 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $9.90 \mu\text{J}$.)

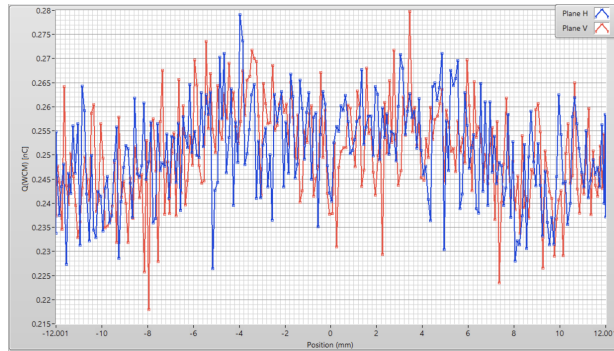


FIGURE 33: Surface QE map of Cathode #210 at 430 nm, with movement on the x-axis shown in blue and on the y-axis in red. (Data was taken with a laser pulse energy of $11.80 \mu\text{J}$.)

Appendix D

TABLE 1: Spectral data for Cathode #210.

Wavelength [nm]	QE Value [%]	Energy [eV]	QE Std Dev [%]
240	7.41	5.17	3.12×10^{-1}
250	8.92	4.96	4.89×10^{-1}
266	10.8	4.66	6.93×10^{-1}
280	12.4	4.43	1.83
290	8.99	4.28	1.52
300	5.29	4.14	4.16×10^{-1}
310	3.12	4.00	5.14×10^{-1}
320	1.67	3.88	9.54×10^{-2}
330	7.78×10^{-1}	3.76	4.86×10^{-2}
340	4.30×10^{-1}	3.65	2.54×10^{-2}
350	1.71×10^{-1}	3.55	1.28×10^{-2}
360	1.10×10^{-1}	3.45	4.35×10^{-3}
370	6.86×10^{-2}	3.35	4.12×10^{-3}
380	3.87×10^{-2}	3.27	6.09×10^{-3}
390	2.22×10^{-2}	3.18	2.77×10^{-3}
400	7.81×10^{-3}	3.10	1.66×10^{-3}
410	2.26×10^{-3}	3.03	4.18×10^{-4}
420	7.11×10^{-4}	2.95	2.02×10^{-4}
430	1.94×10^{-4}	2.89	6.34×10^{-5}

TABLE 2: Spectral data for Cathode #217.

Wavelength [nm]	QE Value [%]	Energy [eV]	QE Std Dev [%]
240	6.53	5.17	1.06
250	6.59	4.96	8.33×10^{-1}
266	5.34	4.66	1.11
280	4.65	4.43	6.09×10^{-1}
290	3.56	4.28	5.32×10^{-1}
300	2.13	4.14	9.20×10^{-2}
310	1.15	4.00	1.33×10^{-1}
320	5.56×10^{-1}	3.88	2.08×10^{-2}
330	1.29×10^{-1}	3.76	6.44×10^{-3}
340	3.82×10^{-2}	3.65	3.32×10^{-3}
350	1.14×10^{-2}	3.55	3.76×10^{-4}
360	4.07×10^{-3}	3.45	2.55×10^{-4}
370	2.44×10^{-3}	3.35	9.32×10^{-5}
380	1.48×10^{-3}	3.27	4.93×10^{-5}
390	6.06×10^{-4}	3.18	5.82×10^{-5}
400	3.71×10^{-4}	3.10	2.62×10^{-5}
410	1.61×10^{-4}	3.03	3.44×10^{-6}
420	1.22×10^{-4}	2.95	1.47×10^{-5}
430	1.15×10^{-4}	2.89	8.95×10^{-6}

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