

Development of novel rectangular photomultiplier tubes for gamma-ray
spectroscopy

by

Jace Beavers

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AN ABSTRACT OF A DISSERTATION

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DOCTOR OF PHILOSOPHY

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KANSAS STATE UNIVERSITY
Manhattan, Kansas

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Abstract

Through the development of phenomenological, stochastic electron-emission models coupled to finite-element multiphysics simulations, two novel rectangular-window photomultiplier tubes (PMTs) for use in gamma-spectroscopy applications have been designed, and various ultra-high vacuum (UHV) fabrication techniques have been explored. The geometries of commercially-produced scintillation spectrometers have historically been driven by standard circular-window PMTs with a maximum diameter of 127 mm, limiting the absolute efficiency and mobility of such devices. There exists a need for rectangular scintillation spectrometers having a large frontal-area and reduced thickness to support discreet, high-efficiency detection and analysis of radioactive materials. Compact, rectangular PMTs and solid-state detectors may be tiled to achieve fine-resolution spectroscopy on such scales. However, increased cost, complexity, and power consumption have limited their implementation in mobile detection applications. Large-area rectangular PMTs circumvent many of these issues, while simultaneously improving photon collection and spectrometer resolution.

To support coupled gamma-ray/optical photon transport simulations used for the development of novel scintillation spectrometers, experiments exploring PMT anode response and the various sources of spectrometer resolution were performed using an ET Enterprises 9490B PMT as a test article. Spatially-dependent relative anode response measurements were taken to understand how response non-uniformity affects spectrometer resolution. Collimated ^{137}Cs gamma-ray beam measurements were used to investigate the spatial dependence of scintillator response for unique crystal geometries. These results will be used to inform and validate Monte Carlo simulations in the future.

Stochastic models of photoelectron emission from the photocathode and secondary electron emission from the dynodes were implemented in COMSOL Multiphysics finite-element simulation software to enable novel PMT design investigations. Coupled electrostatic and

electron-transport simulations were validated using the well-quantified performance of an ET Enterprises 9214B PMT. This methodology was then carried over to an iterative design process to develop and predict the single and multi-photoelectron response of novel PMTs with reflective and transmissive photocathodes.

Finally, several vacuum fabrication techniques were investigated to explore novel PMT fabrication and are discussed herein. A metal vacuum envelope was produced through traditional machining and polishing processes, while additional components were added via laser welding. Two approaches to compact multiplier structure fabrication were investigated. Glass-glass and glass-metal UHV sealing methods are described and testing was performed on borosilicate samples. Finally, a UHV physical vapor deposition and assembly system was constructed for sensitive thin film growth and PMT fabrication. The initial characterization of this system and thin film growth efforts are described. Alternative fabrication methods are also discussed.

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Dedication

To my parents, Pat and Angela, and my wife, Samantha, for their unyielding love, support, and patience. Thank you for all you've given me.

Chapter 1

Motivation and Background

1.1 Introduction

High-sensitivity radiation detection and isotope identification at large standoff distances necessitates the development of large-area, low-power gamma-ray spectrometers with sufficient resolution. Radiation detectors must balance spectroscopic performance, power-consumption, and mobility for any particular application or mission space. For decades, photomultiplier tube (PMT) based scintillator detectors of various shapes and sizes have been adapted for static and mobile detection applications. The design of these devices has largely been driven by commercially available photodetectors, which are often limited by size, performance, or geometry. While material and other technological developments have driven spectrometer resolution downward, photodetector geometry has remained largely unoptimized with regards to its contribution to spectrometer efficiency, resolution, and mobility. The current work seeks to advance spectrometer performance in each of these areas through the development of novel PMT designs which better couple to unique, large-area, rectangular scintillator crystals favored for mobile detection and high sensitivity detection applications.

1.2 Gamma-Ray Interactions in Matter

Gamma rays are high-energy photons emitted from atomic nuclei. Both natural and artificial processes may result in gamma-ray emission. Natural sources of gamma radiation can be of terrestrial or cosmic origins. Cosmic gamma rays may be emitted by astrological sources such as pulsars, quasars, gamma-ray bursts, or solar flares. Additionally, high-energy particles known as cosmic rays may interact in the Earth's atmosphere and produce pair-production gamma rays. Two artificial terrestrial sources of gamma radiation are nuclear fission and fusion⁵.

Terrestrial gamma-ray sources emit high-energy photons following a nuclear reaction, such as fission, fusion, or alpha, beta, and neutron decay. A nuclide left in an excited state following one of these reactions may promptly emit a gamma photon. Other times, the nuclide may reside in an excited state for much longer. This is known as a metastable state. A metastable nuclide, or isomer, spontaneously decays after some period, generally accepted to be greater than 10^{-9} s. The process of isomeric transition (decay) is discrete, but the probability of decay may be described by a continuous distribution. The probability that a transition will occur per unit time is the decay constant, λ , however decay-rate is typically referred to as the time it takes for the radioactivity of a nuclide to be reduced by half. This is known as the half-life ($T_{1/2} = \ln 2/\lambda$)³⁷. After the transition, the isomer may be left in the ground-state, after which it is no longer radioactive barring an induced nuclear reaction. Gamma rays from radionuclide emissions have unique energies corresponding to the specific decay which produced them. Figure 1.1 illustrates the decay scheme of ^{137}Cs . Because of this property, it is possible to identify the source of gamma radiation by analyzing its energy signature. However, identifying a source of gamma radiation is not necessarily a simple process. Due to confounding factors in the absorption and detection of gamma radiation, a distribution of signals is produced. It is, therefore, important to fully understand gamma-ray interactions and signal production to enable effective analyses. The following sections describe gamma-ray interactions in matter and will, hopefully, contextualize the spectral-analysis process.

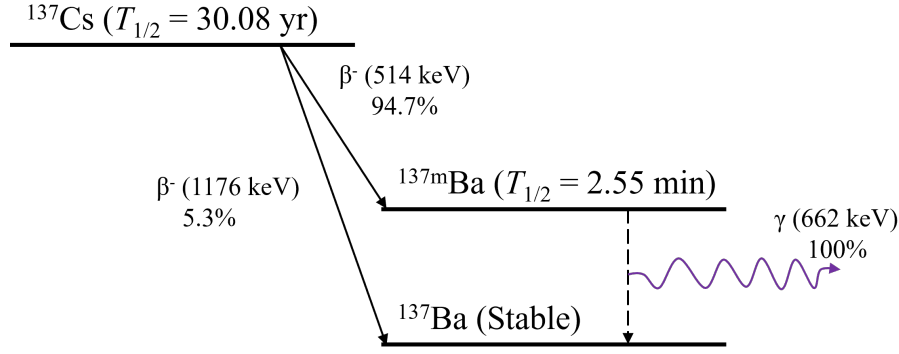


Figure 1.1: The decay scheme of ^{137}Cs as it transitions to stable ^{137}Ba ¹.

1.2.1 Photoelectric Absorption

Photoelectric absorption occurs when a gamma ray interacts with an atom, and the entire energy is transferred to an orbital electron. Nearly all of the impinging photon energy, less the electron binding energy, is converted to kinetic energy in the photoelectron which is either excited into a higher electron shell or liberated³⁷. The vacancy left in the atomic shell is filled by a series of electron transitions to lower shells resulting in the emission of x rays with energies corresponding to the difference binding energies between the shells. The x ray and photoelectron transit the material losing energy through various photon and electron-electron interactions, producing additional liberated electrons and low-energy photons. This process continues until the gamma-ray energy is dispersed, and electron excitation is no longer possible⁵. In this way, all of the original photon's energy is captured. Figure 1.2 illustrates the process of photoelectric absorption by an electron in the atomic shell.

1.2.2 Compton Scatter

Gamma rays may be incoherently or *Compton* scattered when interacting with an individual free or loosely-bound electron. A fraction of the photon energy is imparted to an electron (Fig. 1.3). The energy of this recoil electron is of particular interest to spectroscopic applications because of its contribution to the gamma-ray energy distribution. The energy of this

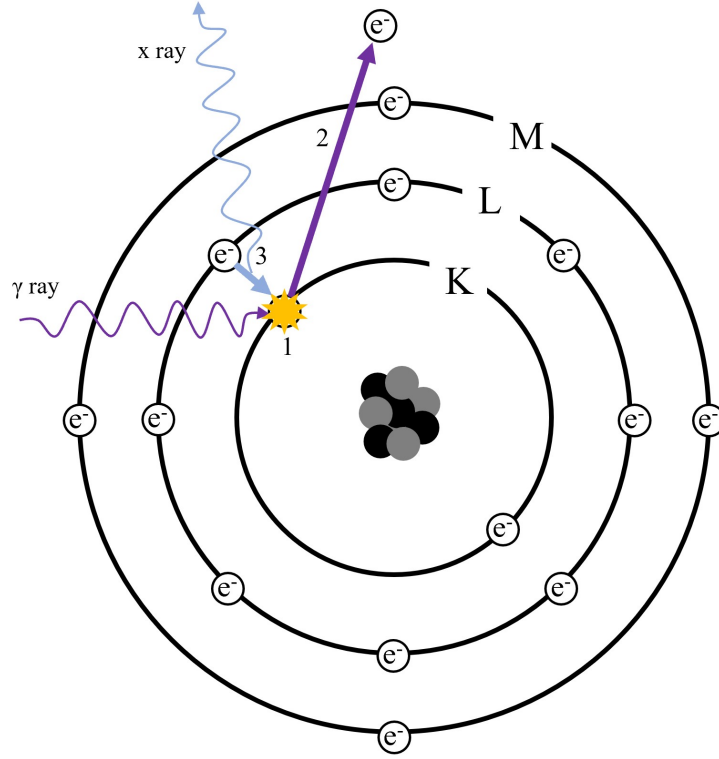


Figure 1.2: *The photoelectric absorption process.*

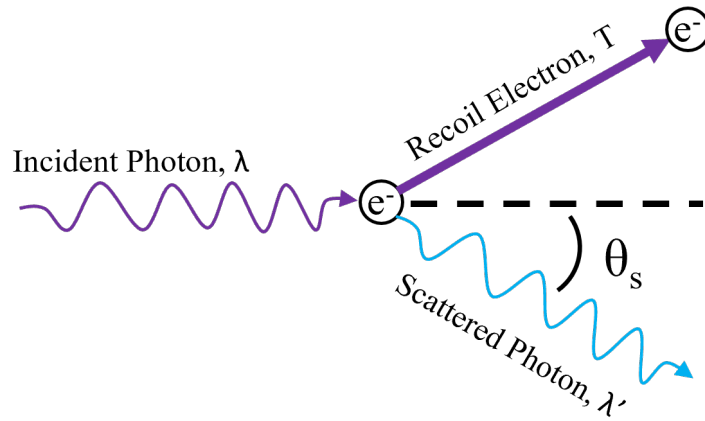


Figure 1.3: *The Compton-scatter process. The differential scatter cross section was first calculated by Klein and Nishina².*

recoiled electron may be calculated using

$$T = \frac{E(1 - \cos \theta_s)}{m_e c^2 / E + (1 - \cos \theta_s)}, \quad (1.1)$$

where E is the initial gamma energy, m_e is the rest mass of an electron, c is the speed of light in a vacuum, and θ_s is the scatter angle referenced from the initial direction of travel. From this, the maximum energy of a recoil electron caused by a 180° scatter angle 662 keV gamma ray from ^{137}Cs is about 478 keV. Oftentimes, the scattered gamma ray escapes the detector medium without further interaction, producing a sharp edge preceding reduced counts between the continuum and full-energy peak. Additional scattering from surrounding materials, known as backscatter, may also contribute to the pulse-height spectrum. A backscatter peak may be produced in an environment where 180° scatter from the surroundings is common. Shallow angle scatters off of the environment may contribute to the spectrum over the entire energy range.

1.2.3 Pair Production

At higher energies (>1022 keV) pair production may occur, producing a prominent spectral feature. During pair production, a gamma ray interacts in the nucleus of an atom directly converting energy into mass in the form of an electron-positron pair. 1022 keV is the minimum threshold energy for this interaction as a positron and electron each have a rest mass of 511 keV. Any excess energy imparted by the gamma-ray is divided between the two subatomic particles as kinetic energy. Both particles move a short distance through the detector, transferring their kinetic energy to other particles along their paths. Eventually, the electron loses enough energy and may rejoin the shell of an atom, while the positron recombines with another electron, annihilates, and produces two 511 keV photons. These photons may be reabsorbed by a detector and contribute to the full-energy peak. Alternatively, one or both photons may escape the detector volume, producing peaks in the distribution at energies 511 keV and 1022 keV below the full energy peak. These are known as the single escape peak and double escape peak, respectively.

1.2.4 Interaction Coefficients

The attenuation of high-energy photons is largely dependent on the atomic number, Z , and the density, ρ , of the material. Although they are independent material properties, a higher atomic number means that the individual atoms within a material are larger, while a higher density means that there is more mass per volume. Increasing one or both of these properties, means that the probability of interaction, i.e. the cross section, is also increased. High-energy photons travel through media in straight lines, disrupted by point interactions as previously described. These interactions occur randomly, thus it is necessary to describe interaction frequency in a probabilistic sense. The linear interaction coefficient, μ_i , for some interaction, i , describes the probability of interaction per unit differential path length for a particle traveling in a given material. Figure 1.4 shows the total interaction coefficients for a few relevant detector materials. The rate of change in the number of uncollided particles,

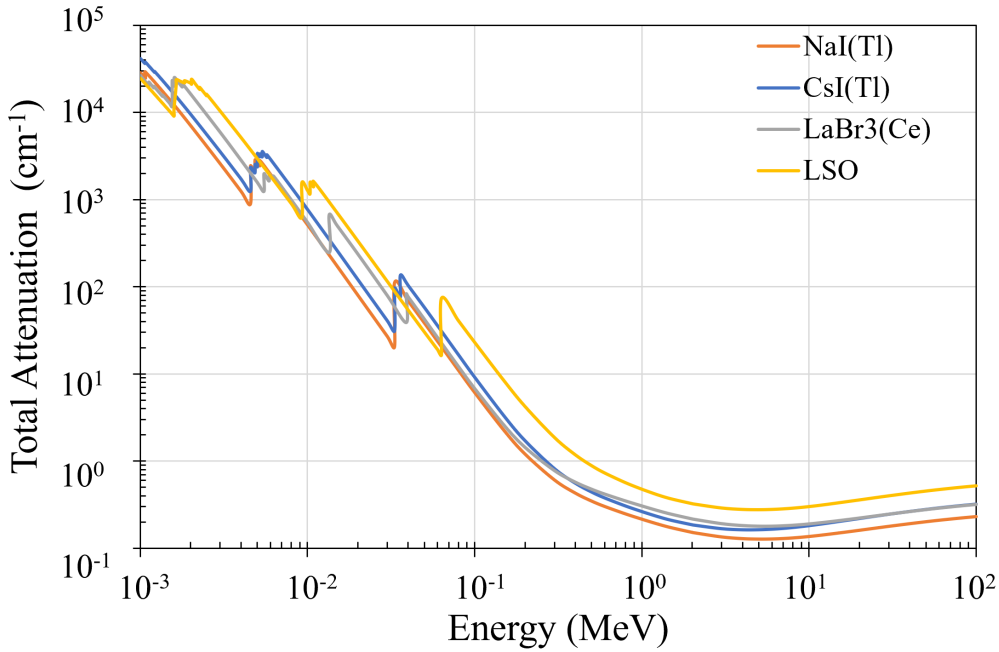


Figure 1.4: The total interaction coefficient for a few inorganic scintillators³. The higher effective Z and density of LSO greatly increases gamma-ray attenuation at energies ≥ 65 keV.

dn , over a differential length through the material, dl , is equivalent to the negative of the linear interaction coefficient times the number of particles, $-\mu_i n$. Rearranging and solving

the differential equation, one finds

$$\int_{N(0)}^{N(x)} \frac{1}{n} dn = \int_0^x -\mu_i dl \Rightarrow \ln N(x) - \ln N(0) = -\mu_i(x - 0) \Rightarrow \frac{N(x)}{N(0)} = e^{-\mu_i x}; \quad (1.2)$$

the ubiquitous exponential decay equation which may be found in numerous scientific fields.

For any interaction type, there is an associated interaction coefficient. Often, the linear interaction coefficient is instead expressed as the mass interaction coefficient, which is simply the ratio of the linear interaction coefficient, μ_i , to the density of the material, ρ . This conversion provides the interaction coefficient as a property of the material, regardless of the density⁵. Figure 1.5 shows mass interaction coefficients for photons in NaI(Tl). Using the

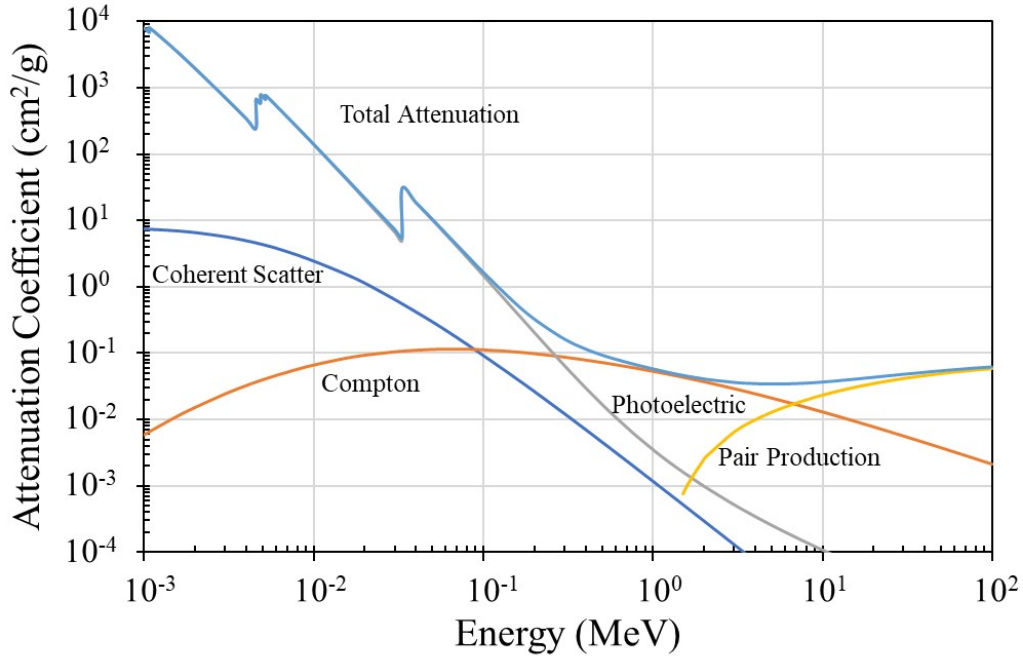


Figure 1.5: *Mass interaction coefficients for NaI(Tl) — a commonly used detector material³. The mass interaction coefficient may also be referred to as the absorption or attenuation coefficient, although not all interactions result in an absorption.*

total interaction coefficient for 662 keV gamma rays in NaI(Tl), one finds that nearly 75% interact within 5 cm.

1.3 Gamma-Ray Detection With Scintillation Spectrometers

Provided a basic understanding of gamma-ray interactions in matter, we may proceed to an overview of gamma-ray detection and spectroscopy using scintillators. Scintillation spectrometers are composed of three primary components:

1. a scintillator to serve as the interaction media which produces scintillation photons,
2. a photodetector to collect and convert the scintillation photons to an electronic signal, and
3. signal-processing electronics (“preamplifier”, amplifier, and multi-channel analyzer or digitizer) to analyze the electronic signal and sort individual pulses into energy bins.

For scintillation spectroscopy applications, inorganic scintillators have been the conventional choice since Hofstadter first developed and tested NaI(Tl) from 1948-1950^{38;39}. The photomultiplier tube is a type of vacuum photodetector; produced for nearly a century following the discovery of secondary-electron emission by Austin and Starke in 1902 and explanation of the photoelectric effect by Einstein in 1905^{40;41}. Slepian created the first secondary-electron amplifier in 1919, a precursor to the photomultiplier tube⁴². PMTs were first conceptualized and developed throughout the 1930s by Kubetsky⁴³, Iams and Slazberg⁴⁴, and Zworkin, Morton, and Malter⁴⁵. Since these historic discoveries, hundreds — even thousands — of scintillator materials and other photodetectors have been produced, each with their own interaction, emission, and signal conversion characteristics.

Inorganic crystals coupled to PMTs have been a mainstay of gamma spectroscopy for their balance of cost, performance, and relative ease-of-use. The absorption of ionizing radiation by a scintillator produces light, with an intensity roughly proportional to the energy deposited by the radiation quanta. This light travels through the crystal and is reflected by the walls of the crystal housing. A fraction of this light may be transmitted through the optical coupling (typically a window and optical coupling grease) and absorbed by the PMT photocathode.

Electrons within the photocathode that absorb a photon have some probability of being excited to a higher energy state and may transit the thin film then be emitted into the photodetector vacuum. These electrons are known as photoelectrons. Photoelectrons are accelerated and focused by an electric field within the photomultiplier tube and are guided to the first secondary-emissive stage, known as a dynode. Photoelectrons impinging the first dynode with sufficient energy may produce a number of secondary electrons. The ratio of the average number of secondary electrons to incident primary electrons is known as total secondary emissive yield (SEY), δ_{tot} . Secondary electrons are again focused and accelerated to subsequent dynode stages, producing a cascade of electrons; amplifying the initial charge and producing a signal at the anode still proportional to the amount of light emitted by the scintillator. Recording signal pulses using a multi-channel analyzer (MCA) or digitizer event-by-event creates a differential pulse height spectrum. This spectrum, effectively a histogram of relative pulse sizes, may be calibrated based on known energies to produce an energy spectrum for further analyses and isotope identification. Figure 1.6 shows an example spectrum for ^{60}Co gamma-rays interacting in NaI(Tl) paired to a PMT.

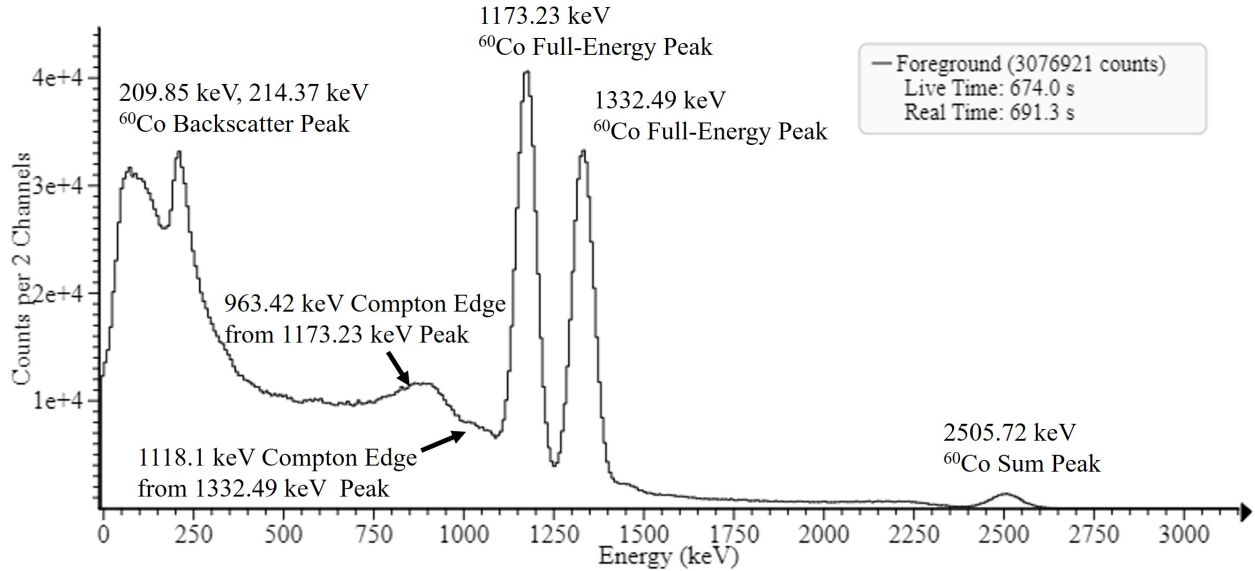


Figure 1.6: A ^{60}Co differential pulse height spectrum recorded using a $2 \times 4 \times 6 \text{ in.}^3$ NaI(Tl) crystal paired to a 2 in. PMT. Note the spectral features created by mechanisms previously described.

A common method employed to identify an isotope using inorganic scintillators is full-

energy peak analysis. If a sufficient proportion of interactions result in the full energy of the incident radiation being absorbed in the crystal — either in a series of partial energy absorptions or a single, full absorption — a peak is formed in the energy spectrum. This peak may be associated with a radioisotope known to produce gamma rays at that specific energy. The shape of this peak is roughly Gaussian with some underlying background, and the standard deviation, or resolution, is typically described in terms of the full-width at half-maximum (FWHM). FWHM dictates the minimum difference required to distinguish individual energies in the spectrum. Energy resolution is a primary motivator in the development of new detection technologies. Solid-state systems requiring cryogenic cooling provide energy resolutions far lower than scintillator spectrometers; however high costs and reduced mobility have limited their adoption in mobile spectroscopy applications. Scintillators are still one of the primary spectrometer technologies used today, and a number of improvements may yet be made to increase both detection efficiency and energy resolution.

1.3.1 Contributions to Scintillation Spectrometer Resolution

Observing the separate processes occurring within a scintillation spectrometer (Fig. 1.7) we see that contributions to energy resolution may largely be reduced to three primary components: intrinsic resolution, R_i , transfer resolution, R_t , and multiplier resolution, R_m . The overall detector resolution, disregarding electronic noise, is described by⁴⁶

$$R^2 = R_i^2 + R_t^2 + R_m^2. \quad (1.3)$$

Naturally, the reduction of any individual term leads to the reduction of the overall resolution.

Intrinsic Resolution, R_i The resolution intrinsic to an inorganic scintillator is dependent on a number of factors. A scintillating material’s growth is rarely perfect, and light output characteristics are highly dependent upon the concentration and distribution of the luminescent centers responsible for light emission, quenching centers where energy is dissipated

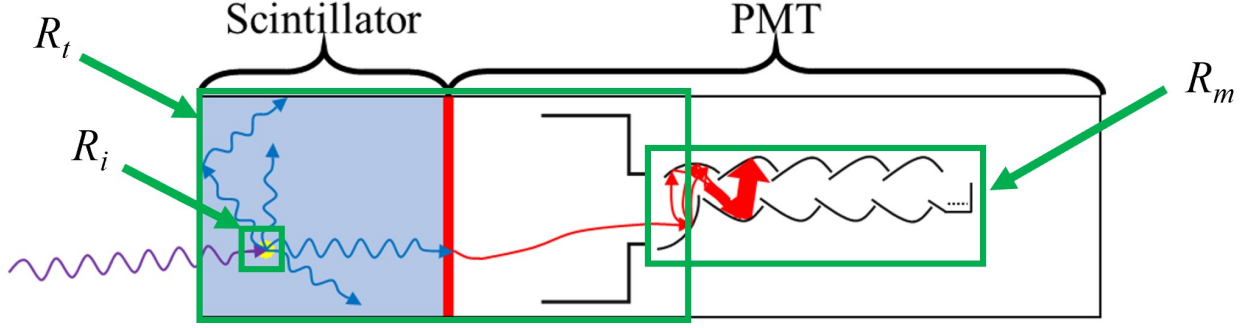


Figure 1.7: *The primary contributions to scintillation spectrometer resolution, disregarding the electronics. A purple line represents an incident gamma or x ray, blue indicates scintillation photons, and red lines illustrate electron transport within a PMT. Green boxes contain the primary regions of interest for the individual contributors to energy resolution.*

thermally, and traps which delay or negate photoemission^{4;47;48}. An uneven distribution of activators can create variance in the number of luminescent centers that may fluoresce, thereby producing spatial nonuniformity in the photon yield. Furthermore, scintillators often display nonlinear or nonproportional behavior, wherein the photon yield for a given energy or interaction is not proportional to that of another energy. This is described in further detail in Section 1.4.2. An approximation of the intrinsic scintillator resolution used in the modern analyses of spectrometer resolution^{46;49}, simplified from Breitenberger⁵⁰, is given by Birks⁴⁷

$$R_i = 2.355 \sqrt{\frac{\text{var}(N_{ph})}{\bar{N}_{ph}^2}}, \quad (1.4)$$

where N_{ph} is the number of photons produced by a scintillation event.

Transfer Resolution, R_t The transfer resolution encompasses all processes from photon emission to photoelectron collection and emission of “useful” secondary electrons at the first dynode. From the point of scintillation, photons are released with an isotropic angular distribution. Each photon will have a different path to the photocathode. Many will undergo one or more reflections, while others may escape or be absorbed. The number of interactions a typical scintillation photon will experience is unique for each spectrometer and is highly dependent upon the reflectors, geometry, and size of the crystal as well as the initial interaction location. The absorption characteristics of a crystal, reflector efficiency and emission

distribution (e.g. diffuse vs. specular), optical coupling, and quantum efficiency distribution over the photocathode each have an effect on the response uniformity, and thus the minimum achievable resolution for a spectrometer. The transfer resolution may be estimated using⁴⁷

$$R_t = 2.355 \sqrt{\frac{\text{var}(\eta_t)}{\bar{\eta}_t^2}}, \quad (1.5)$$

where η_t is the fraction of photons that are converted to photoelectrons and contribute to the signal.

Multiplier Resolution, R_m Ignoring electronic noise, multiplier resolution is the final consideration in the signal-generation process. Although more than 2,000-10,000+ photoelectrons may be produced by the cathode for a full-energy absorption of 662 keV, the variance is larger than the square-root of the number of photoelectrons that Poissonian statistics would suggest, as variance in the single-electron gain of a photomultiplier is inherent to the multiplication process. Additionally, spatial-nonuniformity in the dynode response further increases the signal variance amongst photoelectrons. The divergence of the signal from ideal-Poissonian statistics is described by the Excess Noise Factor (ENF)¹⁸

$$\text{ENF} = 1 + \frac{\text{var}(g)}{\bar{g}^2}, \quad (1.6)$$

where g is the multiplier gain for any individual photoelectron. The overall multiplier resolution is a result of both the number and individual gain of *useful* photoelectrons as follows⁴⁷:

$$R_m = 2.355 \sqrt{\frac{\text{ENF}}{\bar{N}_{pe}}}, \quad (1.7)$$

where $\bar{N}_{pe}$ is the average number of photoelectrons contributing to signal formation.

1.4 Inorganic Scintillators

As previously stated, scintillation is the process by which ionizing radiation energy is converted to photons of relatively low energy. Scintillators are generally separated into two classifications: inorganic and organic. The photoemissive characteristics of inorganic scintillators is largely a product of the electronic band structure of the crystalline material. Within the category of inorganic scintillators, two subcategories, extrinsic and intrinsic, are used to describe materials which may or may not introduce impurities to modify the band structure and, therefore, the emissive properties. NaI(Tl), CsI(Tl), and LaBr₃(Ce) are commonly used scintillators for gamma spectroscopy and high energy physics. These are extrinsic, inorganic scintillators. Impurities, called “activators”, provide additional energy levels within the forbidden band between the valence and conduction bands. These energy levels serve as the luminescent centers which produce scintillation photons that may then be detected.

1.4.1 Scintillation Mechanism

The primary mode of scintillation is described, below^{4;5;51}:

1. Ionizing radiation is absorbed by the scintillator through one of the previously described methods, producing “hot” electron-hole pairs through scattering interactions or by emission of UV and x radiation ($\tau_1 = 10^{-18} - 10^{-9}\text{s}$).
2. Hot electron-hole pairs thermalize, losing energy to the crystal lattice ($\tau_2 = 10^{-16} - 10^{-12}\text{s}$).
3. Thermalized electron-hole pairs form excitons and move to luminescent centers ($\tau_3 = 10^{-12} - 10^{-8}\text{s}$).
4. The excited luminescent centers deexcite via photoemission ($\tau_4 > 10^{-10}\text{s}$).

A simple band diagram (Fig. 1.8) illustrates these processes.

Often, electrons may relax to the ground state non-radiatively. It is useful to look at the single-configurational coordinate (SCC) diagram (Fig. 1.9) to understand how electronic

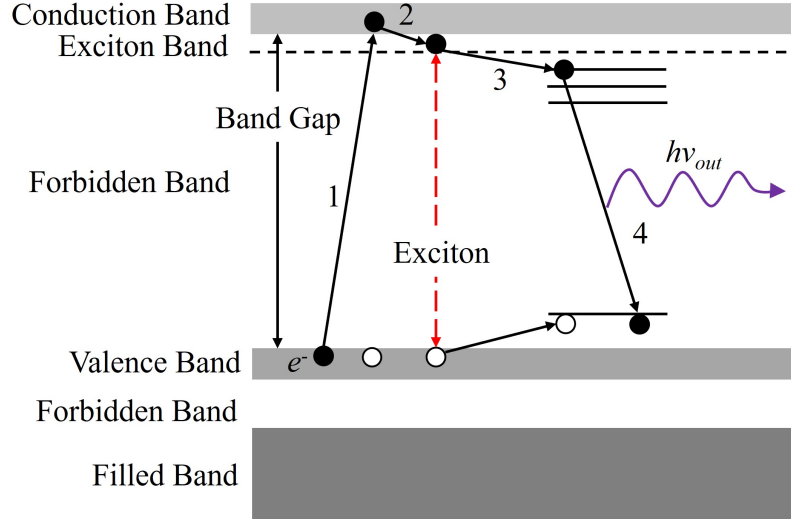


Figure 1.8: *The generalized band diagram and scintillation process for an extrinsic inorganic scintillator.*

states are formed and how electrons move through those states. The potential energy of a spring may be modeled by Hooke's Law,

$$U = kx^2, \quad (1.8)$$

where U is the potential energy stored by a spring, k is the spring constant, and x is the displacement of the spring from its “ground” state. Analogously, a chemical bond between an ion and a luminescent center in a scintillator may be thought of as a spring. For the ground state, the total energy may be described by

$$U_g = k_g Q^2, \quad (1.9)$$

where k_g is the ground-state force constant, U_g is the total energy of the ground state, and Q is the interatomic distance. A similar equation for the excited state is

$$U_e = k_e \frac{(Q - Q_0)^2}{2} + U_0, \quad (1.10)$$

where U_0 is the total energy at $Q = Q_0$. Bonds formed by electrons in the excited state

behave differently than those in the ground state, which means that the force constant, k , is different between states. Figure 1.9 illustrates several electron transitions, due to both radiative (purple) and nonradiative (green) energy transfer.

Consider a photoabsorption for an electron in the ground state ($A \rightarrow B$). The transition occurs almost instantaneously, meaning that the surrounding atomic nuclei appear stationary during the process. In the excited state, the electron occupies a different physical space thereby changing the properties of the chemical bond. Vibrational states produced by thermal motion and the altered bond create a quasicontinuum of energy states represented by horizontal lines. A number of non-radiative (phononic) interactions may relax or excite the

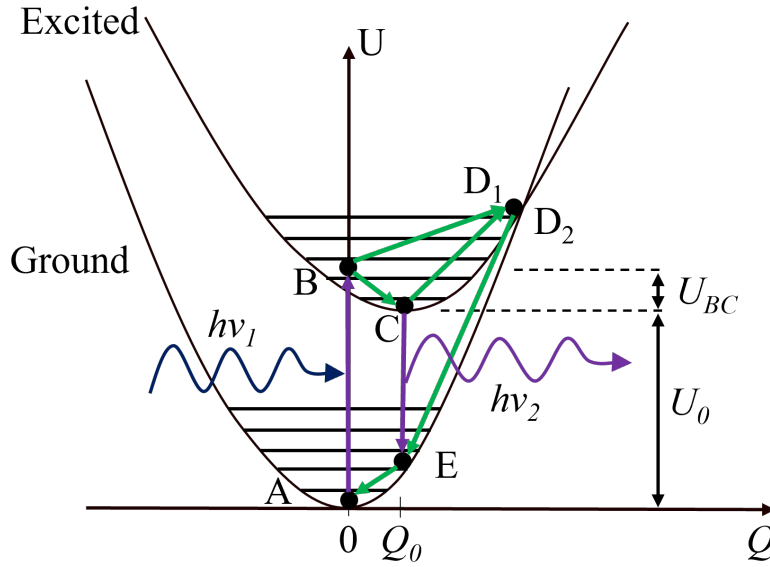


Figure 1.9: The SCC model illustrates the energy states versus lattice vibration for a generalized inorganic scintillator^{4;5}. Purple arrows indicate radiative energy transfer, while green arrows represent non-radiative transitions.

electron within the vibrational bands of the excited state. A non-radiative transfer from $D_1 \rightarrow D_2 \rightarrow E$ is possible since the ground and excited states overlap. An electron may simply transition from the excited state to nearby ground state, passing energy to a phonon. However, $C \rightarrow E$ produces a photon, with $h\nu_2 = U_0 - k_g \frac{(-Q_0)^2}{2}$, which is less than the energy required for photoabsorption. This, of course, is one simple case. A range of transitions may occur throughout the vibrational states, and in fact, they do. Any given transition has an

associated discrete probability, although the density of states is so high, that the distribution appears continuous for any given scintillator. The important detail is that much of the photoemission energy spectrum lies below the energy required for photoabsorption, meaning the scintillator is mostly transparent to its own scintillations. The difference in peak emission and peak absorption, $\lambda_{e,max} - \lambda_{a,max}$, is known as the Stoke's shift, shown in Fig. 1.10.

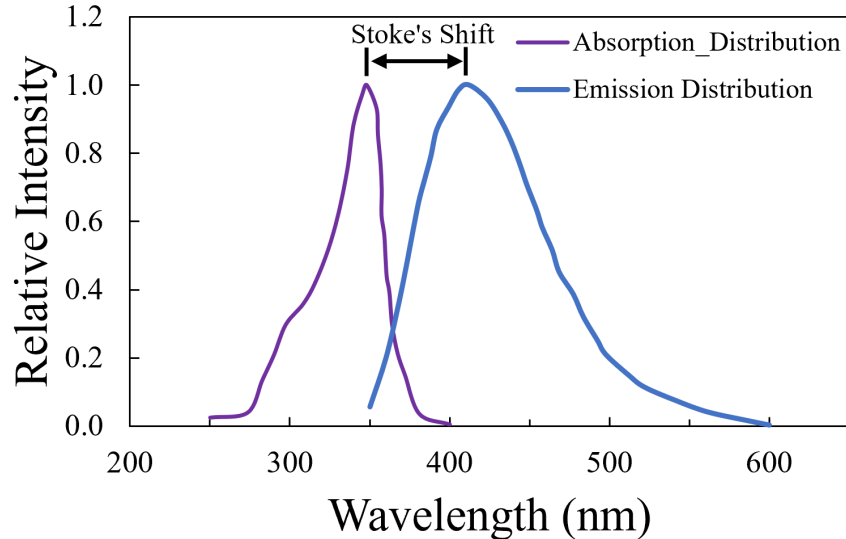


Figure 1.10: *The emission and absorption distributions for NaI(Tl)*⁶.

It is difficult to assign a single intrinsic resolution to a spectrometer, as the detector and source geometries and materials affect interaction type, location, and response. Monte Carlo simulation of a spectrometer may approximate the response and inform design decisions if the scintillator and photodetector characteristics are accurately modeled. The following section provides an overview of the pertinent photon emission and transport properties associated with scintillators.

1.4.2 Light Emission

Light Yield The amount of light emitted by a scintillator is typically reported using the absolute light yield at a specific gamma-ray energy (e.g. 662 keV for ¹³⁷Cs). Absolute light yield is a ratio of the number of photons emitted per unit energy of absorbed radiation, and it is often used to calculate the number of photons produced by ionizing quanta of any

energy. However, absolute light yield may only serve to approximate the average number of photons produced for a specific energy, as inorganic scintillators often display variance in the photon yield per unit energy dependent upon the energy that is absorbed. This is known as nonproportionality or nonlinearity, as the detector response is not linearly proportional over the range of absorbed energies. The absolute light yield for a number of inorganic scintillators may be seen in Table 1.1, while the relative light yield for a few scintillators may be seen in Fig. 1.11.

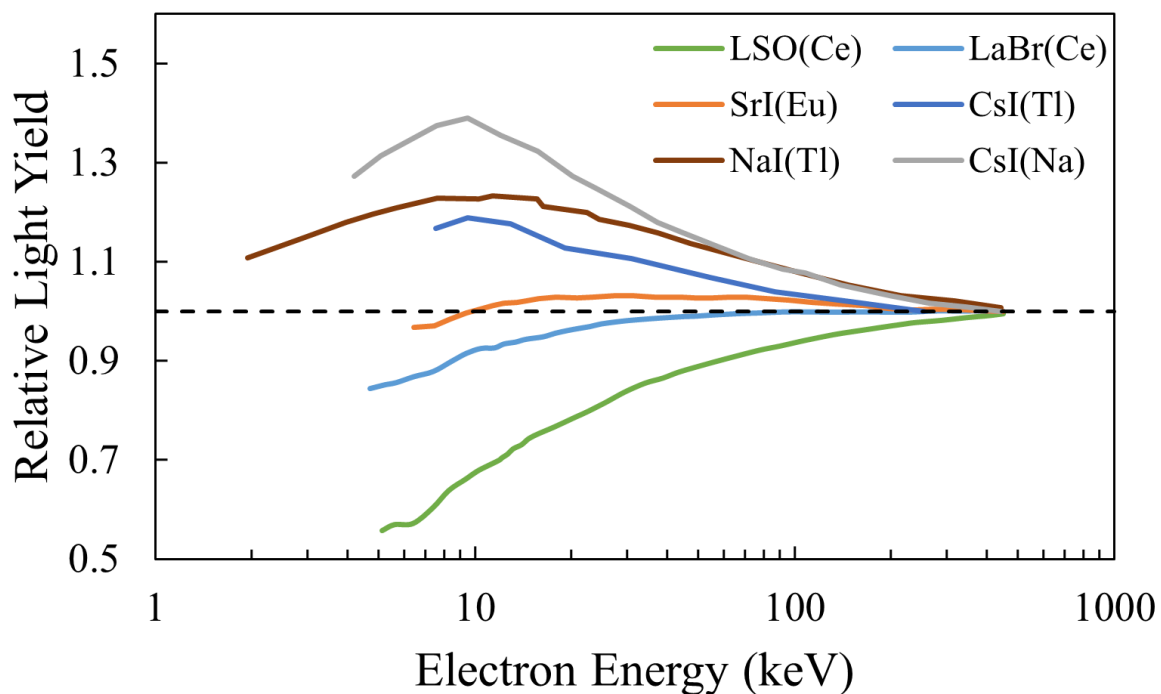


Figure 1.11: *Nonproportionality curves for several scintillators. Data is normalized at 450 keV. Reproduced from Payne and Moses^{7;8}.*

Nonproportionality and photodetector limitations both degrade spectrometer resolution as described by Eq. 1.3. Figure 1.12 illustrates the increase in resolution due to non-ideal scintillation and photon-transport properties. The “ideal” line assumes an ENF of 1.1 and a photon-photoelectron conversion efficiency of 0.25. If scintillators *exclusively* photoelectrically-absorbed gamma rays of any energy, non-linearity would have no effect on detector resolution. However, reviewing the NaI(Tl) interaction coefficients in Fig. 1.5,

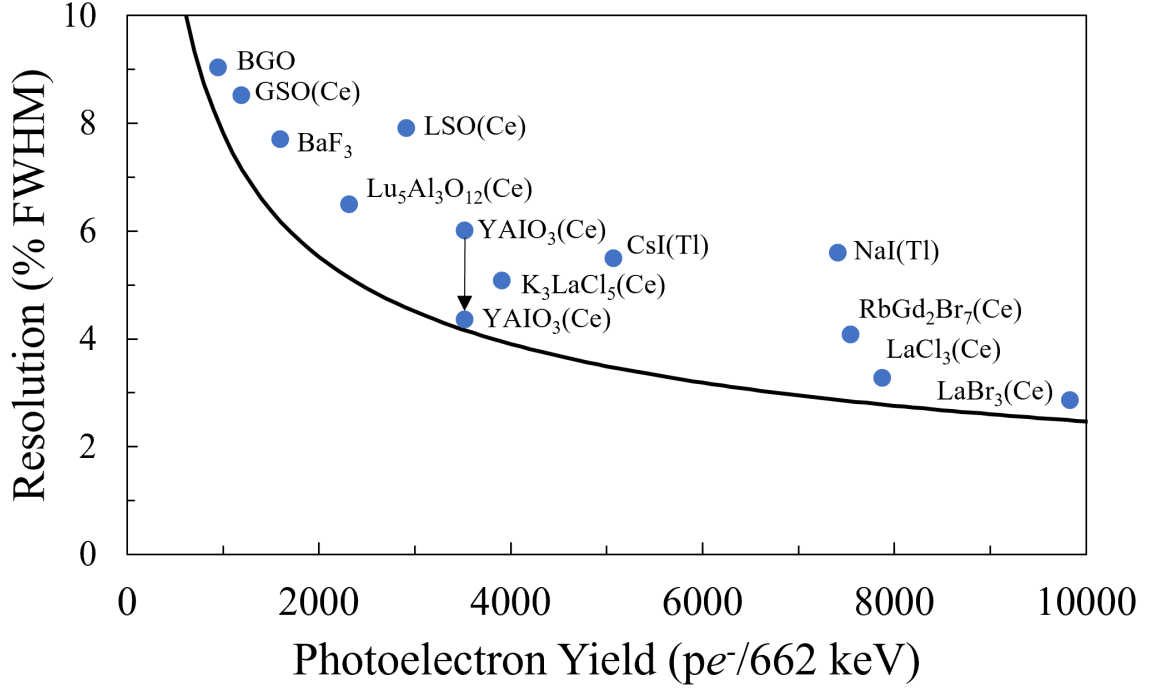


Figure 1.12: Resolution for several different inorganic scintillators compared to the statistically-limited theoretical detector resolution. Reproduced from Dorenbos⁹.

one can see Compton scatter and pair production are the primary interaction processes above 300 keV. Therefore, most full-energy depositions for gamma rays with $E > 300$ keV are a series of scatters punctuated by an absorption, each with an associated relative light yield. The variance in relative light yield depending on absorbed energy means that the total number of photons produced will vary substantially on a per-event basis — beyond the $\sqrt{N}$ standard deviation inherent to Poissonian processes.

Emission Rate The rate of photon emission from a scintillator may be conveniently modeled as the sum of one or more exponential decays

$$\frac{dN(t)}{dt} = N(f_1 e^{-t/\tau_1} + f_2 e^{-t/\tau_2} + \dots + f_n e^{-t/\tau_n}), \quad (1.11)$$

where N is the total number of emitted photons, and f_i is the fraction of photons attributed to the decay constant τ_i . The initial transit of excitons to luminescent centers is generally quite

short and is typically ignored when analyzing the time response of inorganic scintillators. As implied by the inclusion of multiple time constants, the process of light emission is a complex process dependent upon probabilities associated with various absorption, excitation, and emission mechanisms. Scintillators may have differing emission profiles determined by the absorbed radiation, enabling particle differentiation through a technique known as pulse-shape discrimination. For many scintillators, the decay time far exceeds the transit time spread of the photodetector, meaning the pulse shape — and therefore, the maximum count rate — are largely dictated by the scintillator crystal.

Emission Spectra The electronic band structure of a scintillator, which is dependent upon the crystal structure, determines the various emission spectra observed in different scintillators. Figure 1.13, below, illustrates a few emission spectra for common scintillators. It is imperative to ensure that the scintillator, reflective, and optical-coupling materials do

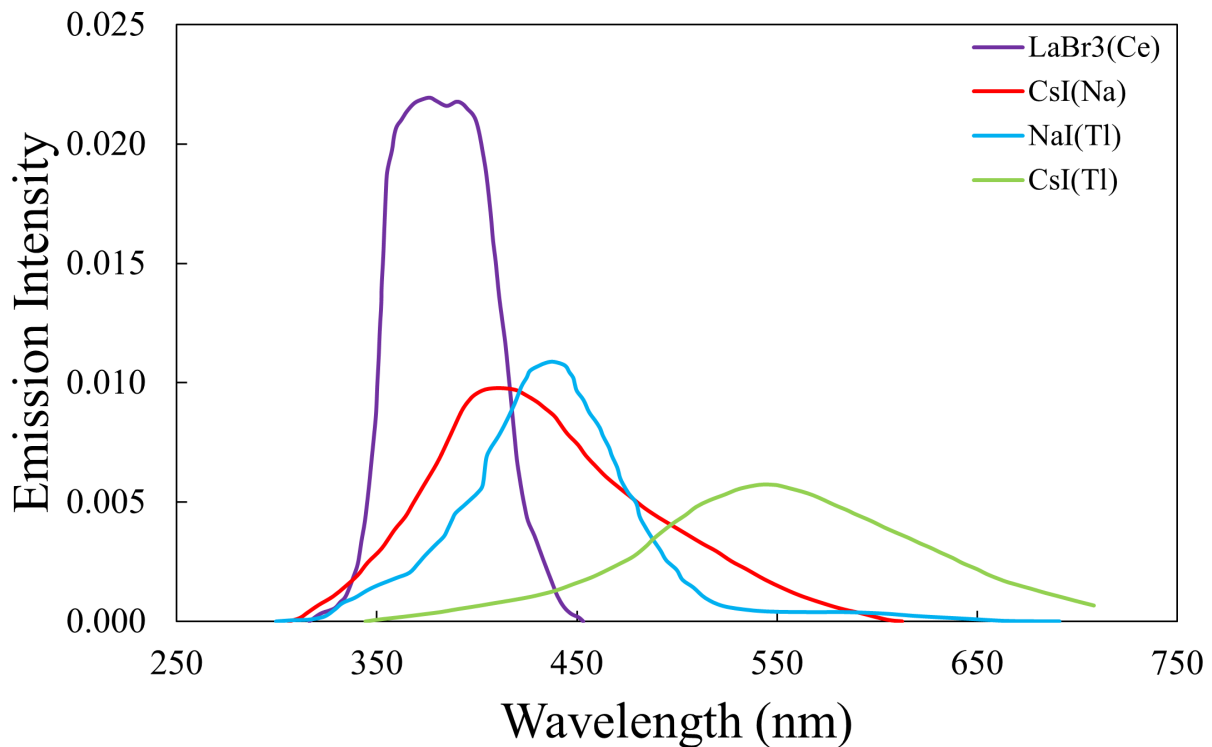


Figure 1.13: The emission spectra for a few inorganic scintillators^{10;11}. The area of each distribution was normalized to one, then scaled by the ratio of their light yield to that of NaI(Tl).

not attenuate the photon signal and that the photodetector has an appropriate detection efficiency over the emission spectrum. A brief review of optical transport and relevant material characteristics are provided in Appendix A.

1.4.3 Inorganic Scintillator Summary

Specific scintillator materials are chosen for a number of reasons (e.g. optical characteristics, emission range, gamma/neutron response, timing characteristics, temperature response, cost), and it's important to choose the material which best suits the application and photodetector. Performance characteristics for some common inorganic scintillators are provided in Table 1.1, below. Rectangular PMT simulations, described in Chapter 3, use the emis-

Table 1.1: *Characteristics of some common scintillators^{10;11}.*

Scintillator	Density (g/cm ³)	Light Yield (photons/MeV)	λ_{max} (nm)	Refractive Index at λ_{max}	Decay Time(s) (ns)	Hygroscopic?
NaI(Tl)	3.67	55,000	415	1.85	230	Yes
CsI(Na)	4.51	41,000	420	1.84	630	Yes
CsI(Tl)	4.51	54,000	550	1.80	600, 3,500	No
BGO	7.13	10,000	480	2.15	60, 300	No
LSO(Ce)	7.40	30,000	420	1.82	40	No
LYSO(Ce)	7.15	25,000	410	1.82	40	No
LaBr ₃ (Ce)	5.20	63,000	380	2.05	25	Yes
CeBr ₃	5.10	60,000	380	2.09	19	Yes
CLYC(Ce) ₃	3.31	20,000	370	1.81	1, 50, 1,000	Yes

sion characteristics of NaI(Tl) due to the material's ubiquity. However, a large focus was placed on compatibility to high performance materials such as LaBr₃Ce which may become the standard if production costs are reduced.

1.5 Photomultiplier Tubes

PMTs are photodetectors used to convert light into an electronic signal. This signal may come in the form of individual pulses from single or multi-photon excitation or as a constant DC signal dependent upon the photon source. From the duration, shape, and amplitude of the electronic signal, information about the light source may be understood. PMT designs are often tailored to specific source characteristics such as timing, intensity, and spatial

distribution. For example, some photomultipliers incorporate plano-concave windows to reduce transit time spread across the photocathode. When paired to fast focus structures and time-compensating dynodes, the light source timing characteristics are effectively maintained. Other PMTs containing continuous multiplier structures offer sub-millimeter spatial resolution and enhanced timing performance at the cost of noise performance¹⁸. By tailoring PMT design to the application space, PMTs have maintained widespread use in scientific photodetection applications such as spectroscopy, diagnostic imaging, and radiation detection.

For many modern general purpose applications such as ambient light sensing, communications, and optical switches, solid-state photodiodes are the preferred choice due to their packaging, affordability, and mechanical robustness, although dark current combined with the lack of internal gain precludes their use in many low-light systems. The development of avalanche photodiode (APD) and silicon photomultiplier (SiPM) technologies have enabled the use of solid-state devices in high-sensitivity applications traditionally reserved for PMTs where compact packaging, low voltage, fast response, and magnetic field insensitivity are required. However, the implementation of SiPM technology is somewhat limited in scintillation spectroscopy due noise and gain performance variability with temperature and comparatively small sensitive areas. Because of this, PMTs are still widely used due to their low light, low noise, and large area detection capabilities. Photomultipliers maintain a number of other advantages over solid state devices including a larger dynamic range and radiation hardness¹⁸. Further development will invariably make a more competitive PMT.

1.5.1 Materials and Construction

PMTs are constructed using high-quality vacuum-compatible materials. The housing or envelope is typically formed of glass or metal. Glass envelopes are chosen based on their structural, chemical, optical, and radioactive properties. Commercially produced transmissive PMTs will often have an inexpensive borosilicate body bonded to another, more expensive window material to take advantage of different optical properties such as UV-transmission,

or index of refraction. Many manufacturers offer low-background glass with reduced ^{40}K content to reduce the radiation background.

Metal housings and internal structures must be nonporous, clean, smooth, and free from high vapor pressure elements such as phosphorous and sulfur. Trapped gasses within cast structures may leak, while rough surfaces and high vapor pressure materials may outgas, providing a continuous source of contamination. Metals commonly used in phototube construction are nickel, BeCu, certain grades of stainless steel, and Kovar — a specialty alloy created for direct glass-metal bonding. Laser-cutting, etching, and forming processes are most commonly employed to form the internal electrode structures. Standard lead-based solders should be used minimally or avoided entirely, therefore most electrical and structural connections are made by spot welding.

To separate high-voltage structures, ceramic insulators are commonly used. Nonpermeable ceramics such as alumina and beryllia may be used for simple structures, while boron nitride and Macor are machinable ceramics adopted to form complex or unique shapes.

Glass-metal bonds for mating optical windows to metal housings or providing electrical feedthroughs may be difficult to produce consistently. Often, metal-oxide-glass bonds such as those formed with Kovar and TiO_2 are sufficient for the small-area bonds formed at the feedthroughs. However, large-area windows bonded to metal housings may employ soft materials to compensate for the mismatched coefficient of thermal expansion (CTE). Indium and indium alloys have relatively low melting/bonding points and are commonly used for UHV chamber windows and other vacuum devices⁵²⁻⁵⁴. It is important to understand the chemical and structural properties of any bond, as In/Au bonds are known to form intermetallics which may become brittle and fail over time^{55;56}.

High performance emissive films in PMTs are extremely sensitive to the internal atmosphere. Common materials are composed of alkali metals, and even a monolayer of O_2 or H_2O will affect the film performance. It's imperative to the growth and long-term viability for the PMT vacuum to contain few reactive contaminants. Contaminants may enter the PMT through a number of means: the initial vacuum may be insufficient to achieve the ultrahigh vacuum (UHV) atmosphere required, seals may fail, small contaminants such as

hydrogen or helium may diffuse through the housing, or contamination present within the building materials may be degassed or desorbed from the surface.

It is important to control each of these sources during production, as the only way to remove them from the atmosphere of a sealed vessel is to use a passive pump known as a getter. Getters come in many forms and many materials (titanium, zirconium, vanadium, etc.)^{57;58}. Getters work by chemically bonding to residual reactive gasses and, thus, cannot pump nonreactive contaminants. Some are evaporable and others are nonevaporable. Depending on the type, contaminants may react at the surface or diffuse into the bulk to form a compound. The type and quantity of getter material depends on the pressure, volume, and makeup of the vacuum atmosphere. PMTs generally employ nonevaporable getters activated by Joule or inductive heating.

1.5.2 Signal Generation Process

Signal generation within a PMT may be separated into four processes:

1. photon absorption and photoemission to vacuum,
2. acceleration and focusing of photoelectrons to the multiplier,
3. electron multiplication and cascade through the multiplier;
4. collection at the anode.

Each process is performed by some component or collection of components within the PMT. Photon absorption and photoelectron emission occurs at the photocathode — the photoemissive material coated on an optical window or substrate within the optical path. Photoelectrons may be emitted over a large area with low energy and must be focused to the multiplier entrance. Focusing electrodes are used to guide and accelerate the photoelectrons towards the electron multiplier.

Two broad categories of multiplier structures commonly employed in PMTs are continuous and discrete. Continuous multipliers consist of a single or collection of channel-like

structures. A sensitive coating impinged by hot electrons will liberate other electrons within the material, yielding some number of secondary electrons per incident electron. This process continues throughout the channel and may generate $\sim 10^4 - 10^7$ electrons per photoelectron. The lack of discrete multiplication stages inherently introduces variance into the signal production process, since the interaction location along the multiplier channel is highly variable. Thus, discrete multipliers are the preferred choice for low-variance applications, such as radiation detection.

1.5.3 Photoemission

The photoelectric effect was first noted in 1887 by Heinrich Hertz⁵⁹. He found that shining an ultraviolet light onto metal plates reduced the voltage at which an arc would form between them. Further investigations regarding the effects of the frequency and intensity of light would culminate in the 1921 Nobel Prize in physics being awarded to Albert Einstein for his description of the of the photoelectric effect in 1905⁴¹. Soon after, the discovery found practical application in phototubes in the form of the AgOCs (S-1) photocathode.

In photomultipliers, the photocathode is typically a semiconducting photoemissive thin film, grown on the window via physical vapor deposition (PVD). Photocathodes are commonly grown on glass, metal, or semiconductor substrates, depending on the application. For photomultipliers, quantum efficiency (QE), describes the ratio of photons “converted” to photoelectrons successfully emitted to vacuum through the photoelectric effect. The Spicer model of photoemission for a common photocathode comprises three parts⁶⁰:

1. Photons are absorbed by an electron within the emitting material.
2. An electron, elevated to a higher energy state within the conduction band, must travel through to the vacuum barrier.
3. Provided sufficient energy, the electron will overcome the vacuum barrier and be released into the vacuum.

Figure 1.14 illustrates this process for a typical p-type, positive electron affinity (PEA) photocathode such as K_2CsSb or Cs_3Sb .

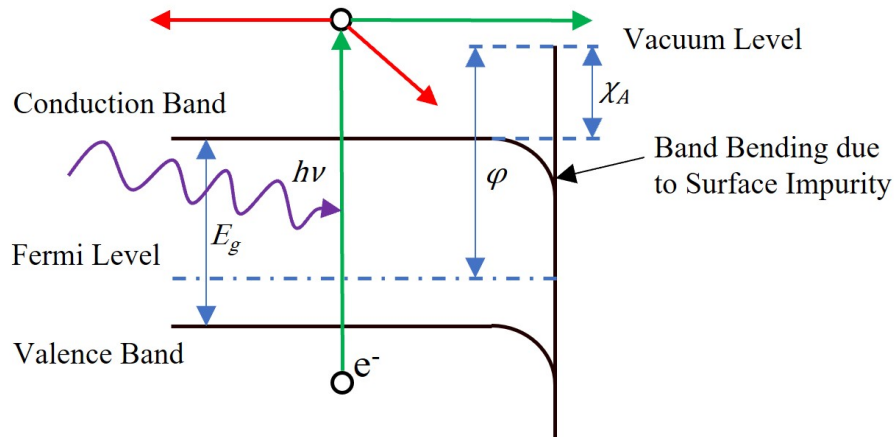


Figure 1.14: The band diagram of a p-type, PEA semiconductor photocathode with band bending caused by surface impurities. Green arrows depict a successful photoemission, while red illustrates an excited electron which does not reach vacuum.

The minimum energy required to move an electron from the top of the valence band to vacuum is called the threshold energy and may be calculated as $E_T = E_g + \chi_A$, where E_g is the band gap and χ_A is the electron affinity. An electron excited by a photon with enough energy to overcome the vacuum barrier may do so if the momentum perpendicular to the barrier is also sufficiently high. Should the excited electron lose energy through various interactions, it may not overcome the vacuum threshold. Similarly, electrons emitted towards the vacuum with sufficient momentum may interact with a phonon, which alters their trajectory such that it is no longer able to overcome the vacuum threshold.

Referring back to the 3-step photoemission process, it is clear that the photoemissive characteristics of a photocathode are dependent upon photon absorption, electron transport, and electron emission into vacuum. A successful photocathode will excel in each of these areas. First, the index of refraction must be well suited to accept photons from the optical interface. The real part of the index of refraction is the ratio of the speed of light in a vacuum to the speed of light in a specific material, $n = c/v$. Snell's Law and the Fresnel equations, outlined in Appendix A, describe the transmission and reflection of light incident upon a boundary of materials with mismatched indices of refraction. A larger discrepancy

in the indices of refraction will enhance both Fresnel and total internal reflection losses at the boundary, reducing the number of photoelectrons produced, and increasing spectrometer resolution. Secondly, the absorption length of a photon in the photocathode should be relatively short, such that electron excitation is confined to a small volume near the vacuum interface to increase photoelectron emission probability. Photon absorption length is described by the complex portion of the refractive index, k , which varies dependent upon the wavelength of incident light, making it hard to tune photocathode thickness to a broad spectrum. The real and complex indices of refraction for a few photocathodes can be seen in Fig. 1.15.

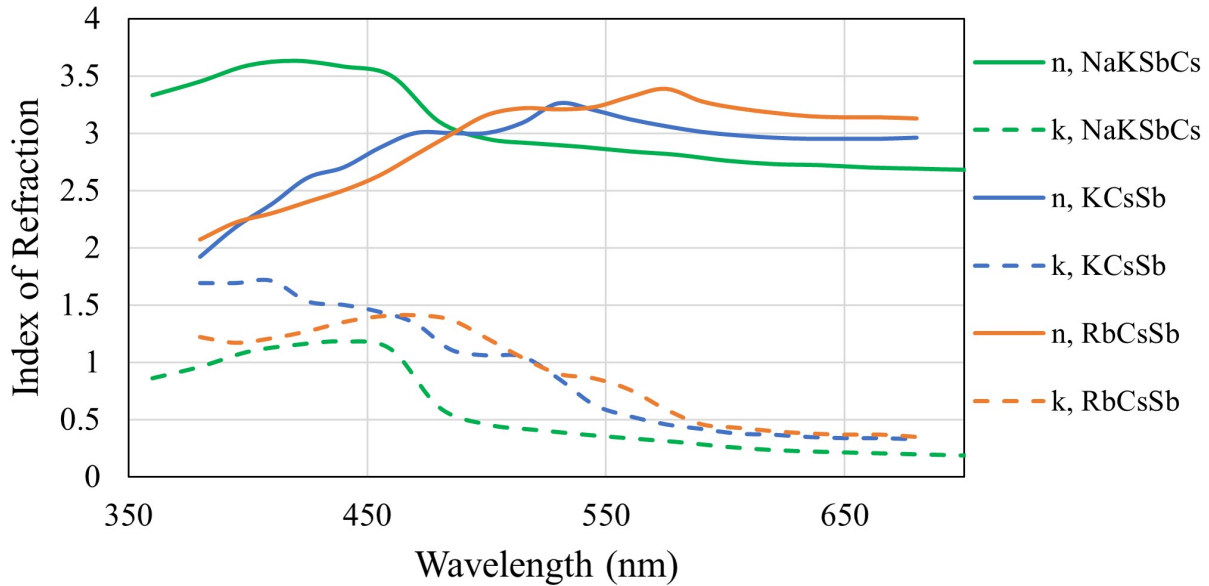


Figure 1.15: The real and complex components of the refractive index for the S-20¹², KCsSb, and RbCsSb¹³ photocathodes are presented.

The photon absorption coefficient may be calculated from the complex index of refraction $\alpha = 4\pi k/\lambda_0$. It may also be calculated with reasonable precision from first principles¹⁵. The absorption coefficient, measured and fit from a first-principles calculation, for a few alkali-antimonide photocathodes may be seen in Fig. 1.16. For a PMT in air with a 20 nm coating of K₂CsSb, nearly 60% of incident 380 nm light may be absorbed by the cathode¹³.

For transmissive photocathode PMTs especially, thickness must be tightly controlled to strike a balance between light absorption and electron transport to the vacuum interface,

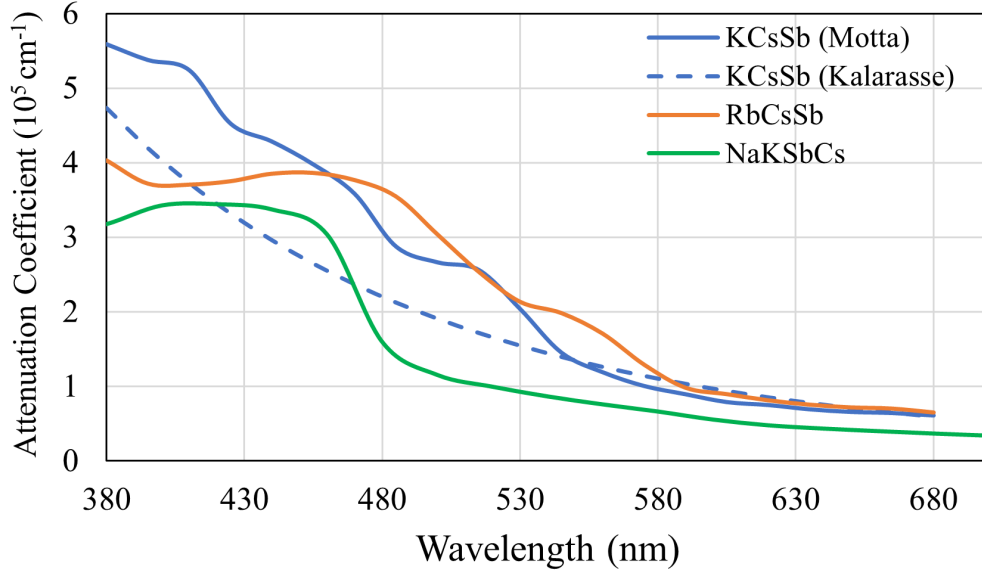


Figure 1.16: *The experimentally derived absorption length (solid lines) for a few photocathodes. A simple quadratic fit¹⁴ (dashed line) to a complex attenuation coefficient distribution¹⁵ is also provided for comparison.*

since photons enter from the boundary opposite of photoelectron emission. In reflective metal photocathodes, the primary photoelectron loss mechanism is electron-electron scattering due to the large number of free electrons. The frequency of these scatter events means that very few photoelectrons — primarily those produced near the surface — will retain enough energy to overcome the vacuum barrier⁶¹. However, in a semiconducting photocathode, because the conduction band is relatively devoid of free electrons and interactions with low-energy electrons in the valence band are forbidden, the primary loss mechanism is phonon scattering. This means that the distance traveled between interactions is quite long in a semiconductor (or insulator), and the emission process is volumetric in nature rather than being confined to the surface. Mean free path (MFP) describes the average distance an electron may travel before an interaction. This value is often considered separately for electron-electron interactions and electron-phonon interactions as the energy loss associated with either interaction type is quite different. The MFP of electron-phonon interactions has been approximated to be 3 nm with an average energy loss of 0.005-0.1 eV in commonly used semiconducting photoemitters, largely impacting photoelectron transport to the vacuum barrier by altering the

momentum^{14;61;62}. Electron-electron interactions typically involve much higher energy losses. The mean free path of such collisions are highly dependent on electron energy and material temperature, as the density of electrons thermally excited into the conduction band residing below the vacuum threshold affects the likelihood of an interaction with photoexcited electrons. Because of the forbidden band present in semiconductors and insulators, there exists a “magic window” in which low-energy electrons are primarily confined to the valence band and a minimum excitation energy of $2E_g$ is required for double-electron excitation.

Photoelectron momentum within the photocathode is mostly isotropic, however due to the perpendicular momentum threshold, electrons are preferentially emitted normal to the surface. The emission-angle distribution is generally considered to be Lambertian, though surface roughness also affects this distribution^{18;63}. Higher extraction gradients at the photocathode surface are often employed to reduce the electron-optical consequences of non-normal emission. The time between photoexcitation and emission to vacuum is typically less than a ps, and not considered to be a major contribution to the overall response time⁶².

Photoemissive materials used in radiation detection applications are chosen based on the properties of the scintillation crystal. The photoemissive characteristics of a few common and historically used materials are provided in Table 1.2. Of course, both the peak and

Table 1.2: *Characteristics of some significant photocathodes*^{16;18;34;35}

Photocathode	Quantum Efficiency at λ_{max} (%)	λ_{max} (nm)	Transparent/Reflective	Features
AgO(Cs)	0.36	740	Either	Earliest Commercial Cathode
Cs ₃ Sb	15-25	340-410	Either	Early High-QE Cathode
K ₂ CsSb	25-45	350-410	Either	High QE, Low Dark Current
Na ₂ KCsSb	15-20	340-440	Either	Improved Red Response
Rb ₂ CsSb	15-25	440-460	Either	Improved Green Response
GaAsP(Cs)	40	540	Reflective	Negative Electron Affinity High QE

spectral QE are important to consider when choosing a photocathode material. In Fig. 1.17, the QE distributions for a few cathodes are plotted alongside common scintillator emission distributions for reference.

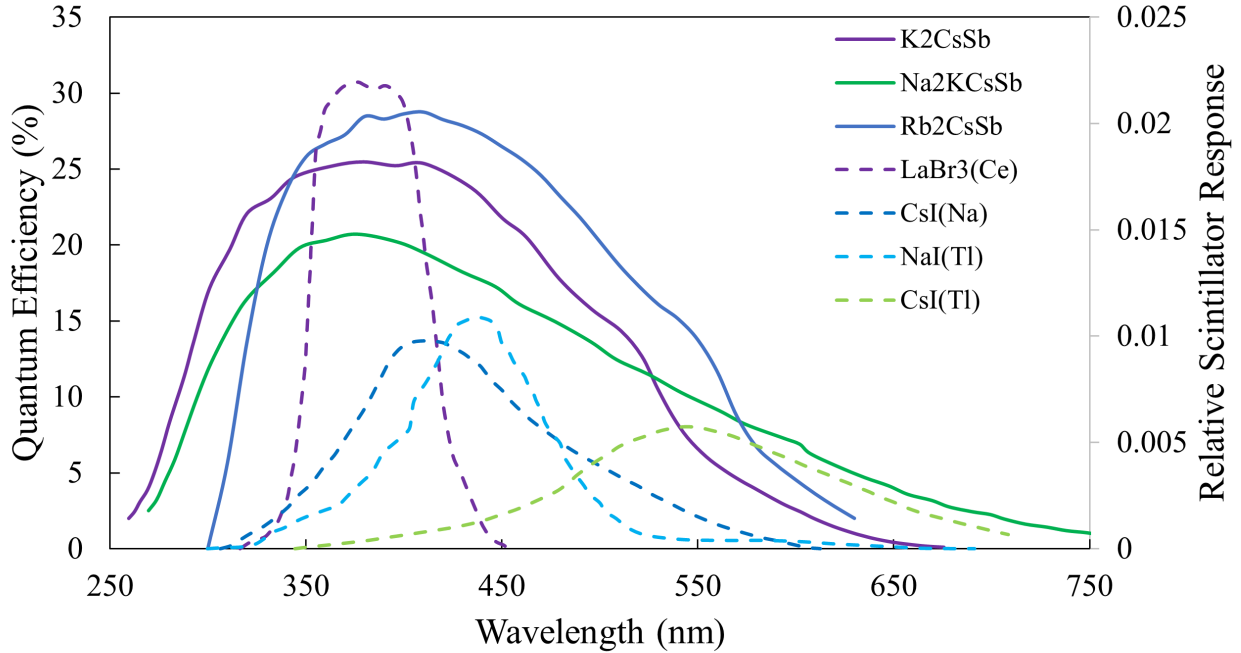


Figure 1.17: Photocathode QE (solid lines) and relative scintillator responses (dashed lines) for a few materials^{10;11;16;17}.

1.5.4 Secondary Emission

Secondary electrons may be emitted by metals, insulators, and semiconductors. However, the electronic structure of these materials greatly affects electron absorption and secondary emission. Secondary emission is a quantum process, but may be qualitatively described in a manner similar to that of photoemission. First, a primary electron penetrates the surface of the emissive material. Next, the energy of the primary electron is distributed to other electrons within the material through a series of inelastic and elastic interactions. Finally, secondary electrons imparted with sufficient energy may diffuse to vacuum.

Electron Interactions In metals, the conduction and valence bands overlap. Therefore, free electrons exist throughout the material and may freely interact with electrons of any energy. The consequence of this being that the MFP of inelastic electron-electron scatters within a metal is very short. Secondary electrons emitted by a metal are excited primarily within the first 5-10 nm of the material, and very few escape — typically $< 2-3$ for relatively

high incident electron energies.

Insulators and semiconductors, however, are defined by the band gap separating the valence and conduction bands. Only free electrons with $E \geq 2E_g$ may interact with those in the valence band. The electron-electron scatter MFP of secondary electrons in these materials is relatively long and another form of energy loss is dominant. Electrons diffusing through crystalline materials may impart vibrational energy to the lattice or reflect off of crystal interfaces. Often the materials used for photoemission may serve as the secondary emitter. This means that the low-energy electron kinetics are the same, and secondary-electron escape-depths of 50-100 nm are possible^{62;64}. Thus, more secondary electrons may escape the bulk of the material.

Electron Emission Energy Spectrum Secondary emission can be described considering three emission modalities:

- A) true secondary electrons (0 - 50 eV),
- B) rediffused primary electrons (0 eV - E_p),
- C) elastically-scattered primary electrons ($\sim E_p$).

Spectral features are the product of unique interaction types, illustrated in Fig. 1.18. I_0 represents the incident current. I_{ts} , I_r , and I_e represent the true secondary, rediffused, and elastic scatter currents, respectively. True secondary electrons are emitted with a relatively low-energy but often in higher number. These secondary electrons are the result of full primary electron energy deposition amongst many electrons in the bulk material. Rediffused primary-electrons undergo fewer inelastic collisions and are reemitted with range of energies spanning the entire distribution. Some true secondary electrons may be emitted alongside a rediffused electron. Elastically-scattered primary-electrons are those that backscatter without depositing a substantial amount of energy. Only one electron is typically emitted as a result. The electron energy distribution associated with each secondary electron emission modality can be seen in the generalized spectrum in Fig. 1.19.

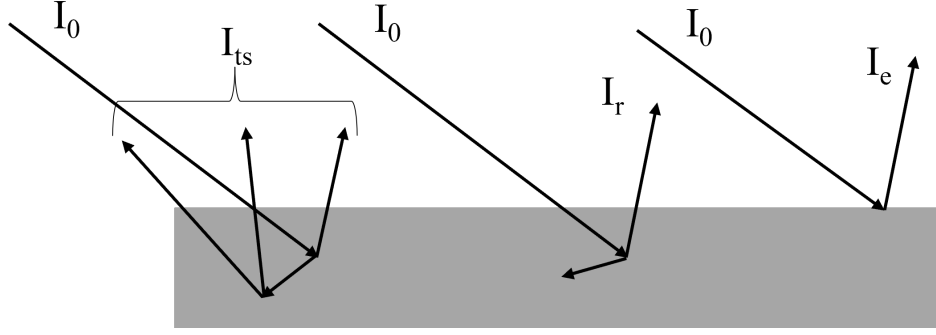


Figure 1.18: Incident electrons, represented by I_0 , generate secondary electrons I_{ts} , I_r , and I_e , each with an associated secondary emissive yield and energy distribution depending on energy deposition and scatter characteristics.

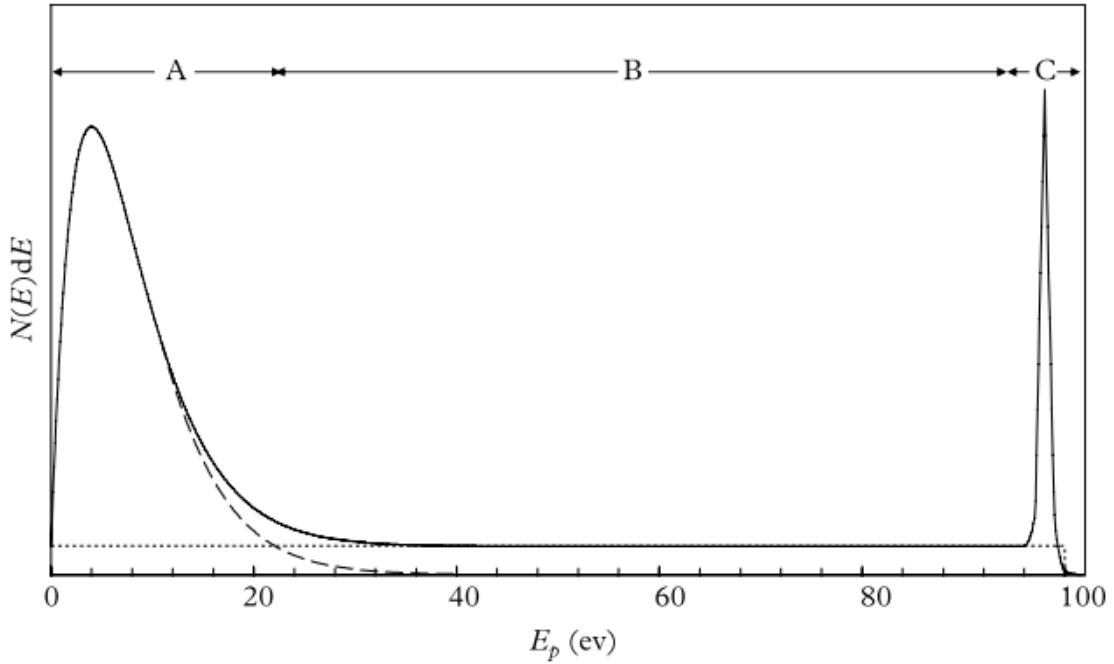


Figure 1.19: A generalized secondary electron energy spectrum, where A, B, and C represent energy ranges over which true secondary, rediffused, and elastically scattered electrons, respectively, are the primary constituents of the distribution¹⁸. Dashed lines indicate the underlying distributions which contribute to the total energy distribution represented by the solid line.

Electron Emission Yield The average number of secondary electrons produced per incident primary electron is known as the secondary emissive yield (SEY). SEY, also represented by δ , has been measured for a number of metals, insulators, and semiconductors^{20;21;65}. The SEY is dependent upon the range and energy deposition profile of the primary electron in

relation to the secondary electron range within the emissive material, as discussed previously. Baroody was the first to discover that plotting the ratio of the energy-dependent SEY and the maximum SEY, $\delta(E_p)/\delta_{max}$, against the ratio of the primary electron energy and primary electron energy at the maximum SEY, $E_p/E_{p,max}$, produced a similar result for most materials. Expanding upon this theory, a number of researchers developed “standard” empirical models of SEY, for which only a few parameters must be changed to effectively map yield over a large energy range^{66–68}. The “constant loss” model of SEY seems to best accommodate for scattering within the emissive material (Fig. 1.20), and may be calculated using

$$\delta = K_m E_p \left[\frac{1 - e^{-\alpha C_m E_p^{n+1}}}{-\alpha C_m E_p^{n+1}} \right], \quad (1.12)$$

where K_m and C_m are constants, α is the electron attenuation coefficient, and n is a fit parameter. The normalized dependence, δ/δ_m , may be found by defining a variable $z \equiv \alpha C_m E_p^{n+1}$, such that SEY is proportional to

$$f_n(z) \equiv \frac{1 - \exp - z^{n+1}}{z^n}. \quad (1.13)$$

The normalized SEY may be written as

$$\frac{\delta}{\delta_m} = \frac{f_n(z_m E_p/E_{pm})}{f_n(z_m)}, \quad (1.14)$$

where δ_m , E_m , and z_m are easily fit to SEY data for a given material⁵. One may, alternatively, simply fit secondary emission data using a polynomial equation if the pertinent energy range is sufficiently encapsulated. Further consideration of backscatter processes and incidence angle must be included for a full model of secondary emission. This will be further discussed in 3.2.4.

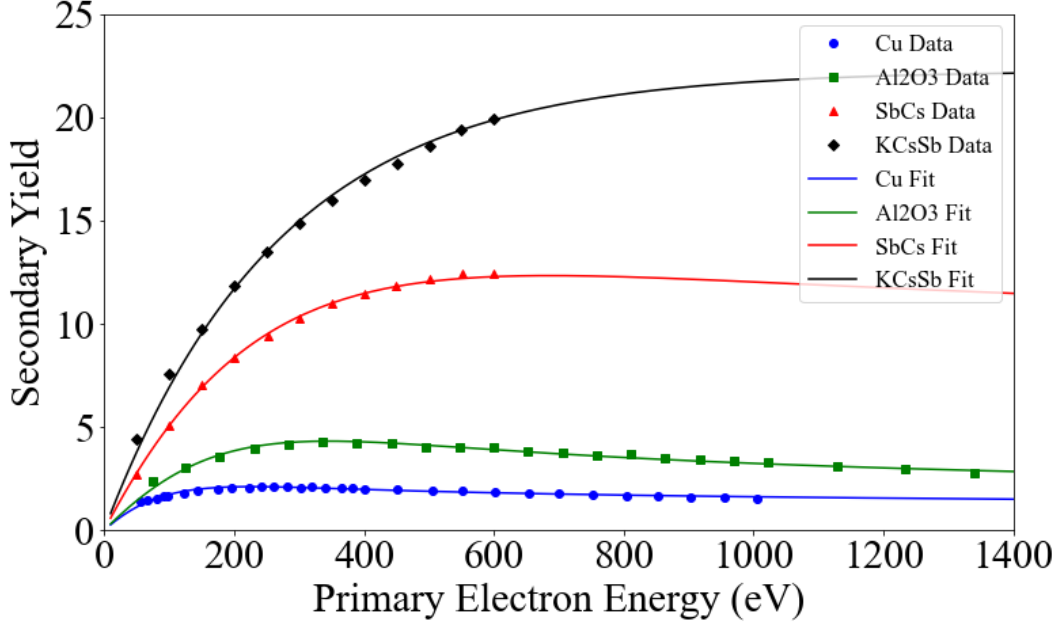


Figure 1.20: *Secondary emissive yield for a few materials fit with the constant loss model. Data are from Sommer, Wei, and Furman for the alkali-antimonides, ALD Al_2O_3 , and Cu, respectively^{19–21}*

1.5.5 Single Electron Response

Energetic electrons impinge the secondary emissive material with a continuous distribution of angles and energies dependent upon the device’s electron optical characteristics. Furthermore, subsurface interactions are stochastic, meaning there is a discrete probability distribution in the number of emitted secondary electrons for any incident electron momentum. Variance in the signal produced on a per-event basis is a source of increased multiplier resolution, R_m . The distribution of charge collected at the anode resulting from single photoelectron emissions, known as the single electron response (SER), may be used to quantify the ENF — an important performance metric for photodetectors used in gamma-ray spectrometers. For continuous multipliers, such as the Kubetsky tube or microchannel plate (MCP), the first secondary emissive event may occur in a range of locations. The ultimate gain of these devices is highly dependent upon the first few interaction locations, creating a wide distribution of single electron responses. Discrete multiplier designs mitigate this effect by guiding electrons to discrete multiplication stages, hence the name. By accurately con-

trolling the location and number of interactions, one may remove a large source of variance contributing to the SER.

For a PMT with an effective photoelectron collection efficiency (the ratio of emitted photoelectrons to those which contribute to the signal at the anode), η_{eff} , an average yield, δ , and m dynode stages, it is common to use

$$G = \eta_{eff} \delta^m \quad (1.15)$$

to calculate multiplier gain. However, this value is only an estimate of the average signal produced at the anode of a PMT and does not consider the variance of that signal which contributes to overall spectrometer resolution. As such, it is important to analyze the statistics of signal formation in a PMT to understand what properties are necessary to achieve low energy resolution. Appendix B provides a full statistical analysis of multiplier noise assuming Poissonian secondary emissive yield. To summarize, the variance at the k^{th} dynode of a multiplier with equivalent SEY, δ , at each stage, and an expected gain, $\langle g \rangle = \delta^k$, is described by $\text{var}_k = \langle g \rangle^2 \frac{1}{(\delta-1)}$.

Often, a higher voltage is applied between the photocathode and first dynode to improve signal timing and gain performance. Given the increased potential difference, the SEY of the first stage, δ_1 , must be considered separately such that

$$\frac{\text{var}_k}{\langle g \rangle^2} = \frac{\delta}{\delta_1(\delta-1)}. \quad (1.16)$$

Observing Eq. 1.16, it is clear that increasing the first-stage SEY reduces the fractional variance, and thus the ENF, thereby improving detector resolution. This strategy is commonly leveraged to reduce the resolution of commercial PMTs. Mathematical analyses such as this provide general guidance on multiplier design and material decisions, however design studies using charged particle tracking simulations must be performed to understand the nuances of photoelectron and interstage collection efficiencies, as well as the additional variance incurred by electron emission characteristics.

1.5.6 Discrete Dynode Designs

As discussed previously, discrete dynodes with high collection, low ENF, and low single-electron resolution are necessary for scintillation spectrometers. Timing characteristics may place an upper limit on count rate, although light collection devices often outpace the standard pulse-processing electronics. Gain may be arbitrarily set, as it is a function of operating potential and the number of multiplication stages. However, space charge accumulated near the anode must be considered for relatively large pulses at high gain, such as those produced by a scintillator. To combat this, manufacturers will often place a wire-mesh anode between the penultimate and final dynode to improve charge extraction, thereby increasing linearity. A number of discrete dynode designs exist; each having strengths and limitations. The cross-sectional geometries of several discrete multiplier design are shown in Fig. 1.21, and an outline of performance characteristics for a few dynode geometries is provided in Table 1.3.

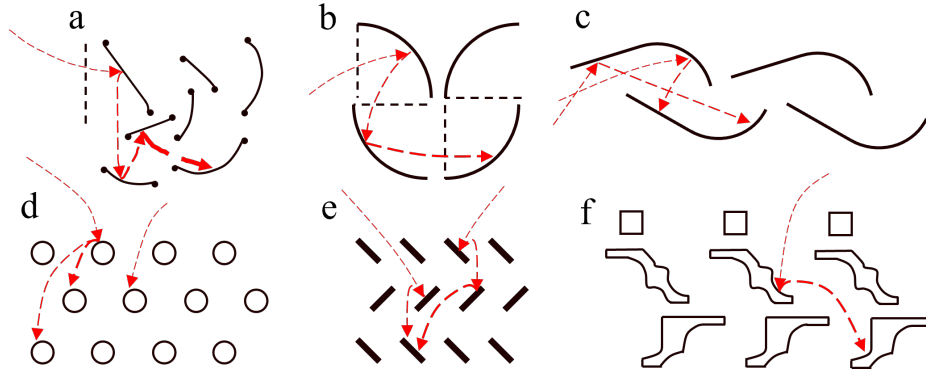


Figure 1.21: *The cross-sections of several dynode geometries, including: a) circular cage, b) box-and-grid, c) linear focusing, d) fine mesh, e) venetian blind, and f) metal channel. Designs are not to scale. Feature sizes for a, b, and c are much larger than d, e, and f.*

Circular-Cage and Linear-Focused Dynodes Some of the first discrete multipliers produced commercially were of the circular-cage design. These dynodes are relatively compact and utilize a compensating electron-optic design to reduce the pulse width. Linear-focused dynodes are quite similar to their circular-cage predecessor. Specially designed electrodes steer electrons such a way to be time-compensating. Figure 1.22 illustrates how secondary

Table 1.3: *Typical performance characteristics for discrete dynode designs*^{16;18;36}.

Dynode Type	Rise Time (ns)	Single pe^- FWHM (ns)	Collection Efficiency	Single-Electron Resolution	Response Uniformity	Notes
Circular-Cage	0.9-3.5	7	Good	Good	Acceptable	Semi-compact
Linear-Focused	0.7-3.0	1.3-5.0	Good	Good	Acceptable	Linear
Box-and-Grid	6.0-20	13-20	Very Good	Good	Good	Simple design
Fine Mesh	1.5-5.5	5.0	Very Good	Very Poor	Good	Linear, Simple fabrication
Metal Channel	0.65-1.5	1.5-3.0	Good	Good	Poor-Good	Compact, Difficult fabrication
Venetian Blind	6.0-18	25	Good	Very Poor	Poor-Good	Compact, Simple design

electrons released closest to the target dynode impact “higher up” the electrode. Conversely, electrons emitted furthest from the target electrode travel a shorter distance to the top of the subsequent dynode. Overall, signal dispersion or “transit time spread” (TTS) is reduced. Strong electric fields at the dynode surfaces ensure efficient electron extraction and improve linearity which would otherwise suffer due to space-charge buildup^{16;18;69}.

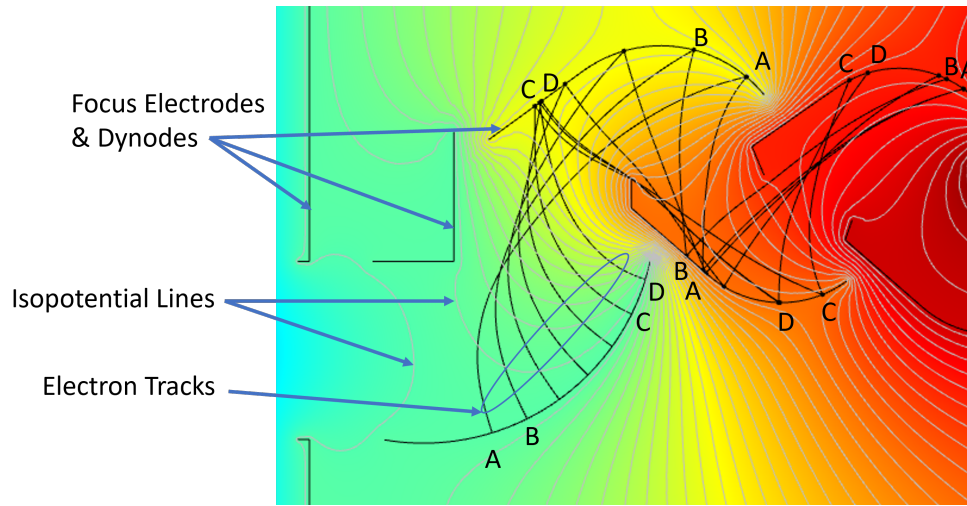


Figure 1.22: A 2-dimensional simulation of electron transport following emission normal to the dynode surface shows how the linear focus design compensates for the emission location by alternating short and long electron transit lengths between dynodes. Black lines indicate electron transit with primary and secondary electron paths denoted by letters A-D, while gray lines represent the isopotential lines of the electric field shown in the colormap. Cooler colors represent lower potentials, while warmer colors indicate higher potentials.

Box-and-Grid Dynodes Box-and-grid dynodes are used prolifically in transmission photocathode PMTs. The simple design utilizes focusing grids at each stage to accelerate sec-

ondary electrons to the dynode surface. The collection efficiency is near 100% at the first dynode and between stages, which produces a well-defined peak in the SER. However, the potential gradient over each dynode surface is relatively weak and inconsistent meaning the initial momentum of a secondary electron has a greater impact on transit time performance. This greatly increases the temporal dispersion and pulse FWHM^{16;18;35}.

Venetian Blind Dynodes Venetian blind dynodes feature a simple, compact parallel-plate construction. Collection at the first dynode stage is relatively high, however the effects of streaming (electrons skipping the next multiplier stage) and low extraction fields mean that these dynodes are more likely to exhibit a quasi-exponential SER and slow timing characteristics. Additionally, venetian blind dynodes may display directional dependence where electrons released from one side of the photocathode are much more likely to skip the first dynode stage. Some manufacturers have avoided this by incorporating a focusing grid or radially symmetric designs¹⁸.

Fine Mesh Dynodes Fine mesh dynodes are exceptionally easy to produce. Wires on the order of 30-100 μm with grid pitch on the order of 1 mm form a cross-hatch pattern. The tight photocathode-dynode 1 and interdynode spacing ensures a high electric field gradient. This high gradient enables high linearity, good magnetic field immunity, and reasonably fast timing^{16;36}. However, poor secondary electron extraction from the top of the dynode requires more multiplication stages (>15) to achieve typical multiplier gains.

Metal Channel Dynodes Metal channel dynodes, as their namesake implies, are a modern design made from thin sheets of metal which are micromachined to achieve a repeating micro-scale linear-focus-like dynode structure. The thin plates are stacked in close proximity to enhance the extraction field, thereby improving timing, collection efficiency, and linearity. These structures often span the full area of 25-51 mm square photodetectors, producing a spatially uniform response if used in conjunction with a focusing grid, as shown in Fig. 1.21. The short photocathode-dynode and interstage distances also reduce spatial dispersion and

retains performance in a magnetic field^{16;18}. However, the micro-scale dynode geometry are more difficult to produce than mesh or large-scale alternatives.

1.6 Proposed PMT Design Improvements for High-Efficiency Scintillators

Common circular-window PMTs pair well, dimensionally, to right-cylinder scintillators, and combined, these detectors provide a relatively uniform response throughout the crystal volume. However, to maximize sensitivity to radiation sources at standoff distances, it is beneficial to increase the detector's frontal area. Cylindrical PMTs mated to rectangular scintillators, such as the example in Fig. 1.23, are not efficiently optically-coupled, since much of the active crystal volume is not directly visible to the photodetector. Indirect pho-

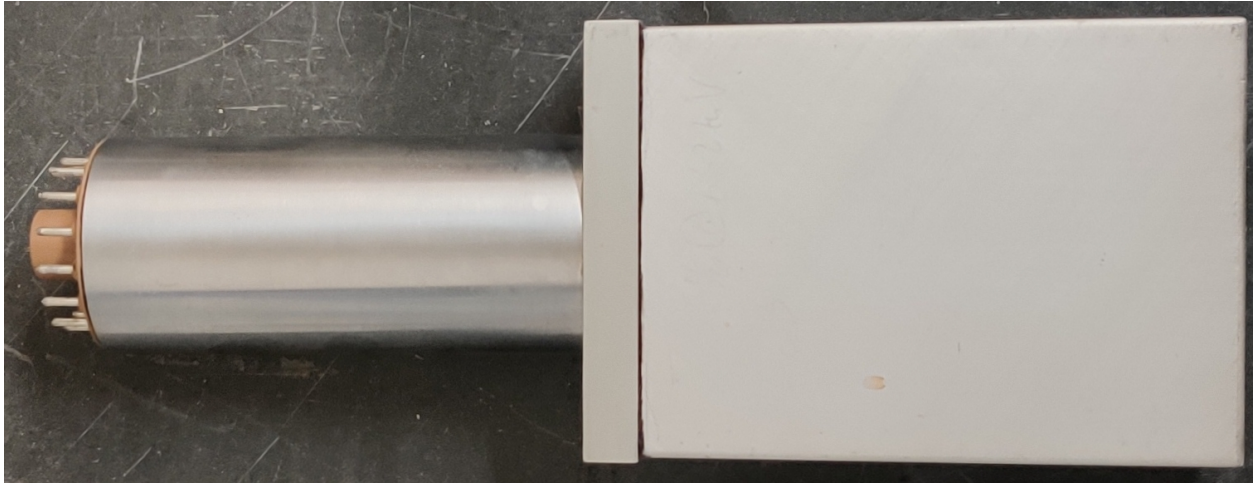


Figure 1.23: A $50.8 \times 101.6 \times 152.4 \text{ mm}^3$ crystal mated to a standard 50.8 mm diameter PMT such as this one leaves nearly 61% of the optical boundary uncovered. Scintillations originating in the corners of the crystal adjacent to the PMT will have a disproportionately small response, increasing the spectral resolution.

ton transport to the photocathode increases the chances of optical photon losses through boundary interactions and self-absorption. Any reduction in the total number of optical photons collected magnifies the effect any single photon has on the signal magnitude which increases fractional variance, and thus, the energy resolution. One can easily understand

this relationship through Eqs. 1.5 and 1.7. The proposed solution matches the PMT optical window and photocathode geometries to that of a rectangular scintillation crystal thereby enhancing optical transfer efficiency and spatial uniformity.

Another deficiency of the standard PMT design is the size required to accommodate focusing structures as well as 8-12 dynode stages. The primary advantage of using high-gain PMTs is low-noise signal amplification. PMTs can provide gains of 10^5 - 10^7 electrons per photoelectron, enabling standard electronics to detect single-photoelectron induced signals. Part of the proposed solution instead relies on the significant photon emission produced via scintillation to reduce the photodetector gain-burden and size. Additional signal amplification may be achieved with standard electronics used for gamma-ray spectroscopy, while higher sensitivity is realized by adapting charge-sensitive amplifiers traditionally found in solid state and gas detector applications.

Following this hypothesis, the Integrated Circuits and Systems Lab at the University of Nebraska-Lincoln developed a small-scale spectrometer composed of a Hamamatsu R11265U PMT operating with just four multiplication stages at 500 V paired to a $\text{LaBr}_3(\text{Ce})$ scintillator (Fig. 1.24). The spectrometer achieved a 3.0% energy resolution at 5 keps, while

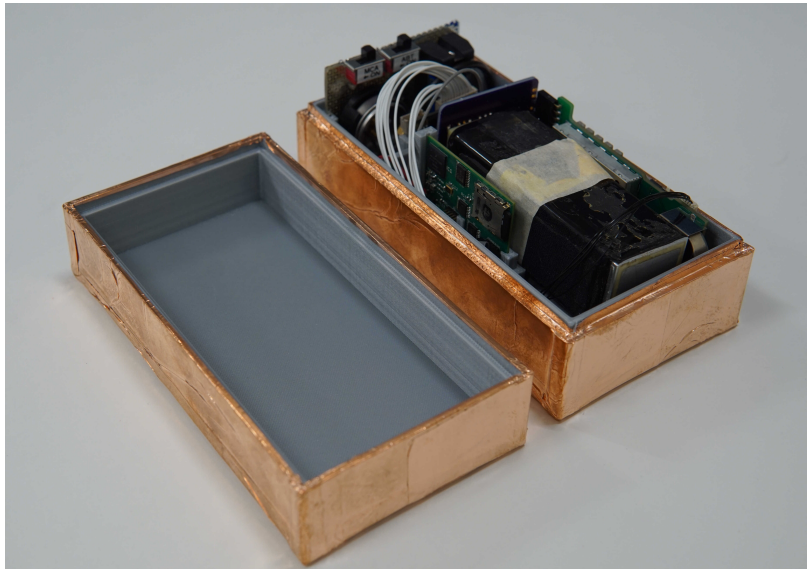


Figure 1.24: The 4-stage $\text{LaBr}_3(\text{Ce})$ spectrometer. Photo courtesy of the Integrated Circuits and Systems Lab²².

consuming less than 21 mW of power by utilizing an efficient power supply, sensitive preamplifier, and optimized background anomaly detection algorithm²².

It is believed that a spectrometer of comparable performance may be built upon a large-crystal platform by leveraging an optimized photodetector geometry. The optimized geometry will have a rectangular window and extended, miniaturized dynode stage to reduce signal variance along the photodetector length. An offset reflective photocathode is proposed to distribute photons and improve response uniformity. The reflective design should have a small, discrete dynode assembly atop the window to limit non-productive photon absorption by multiplication and focus structures. Initial designs incorporate a flat, optics-grade borosilicate, though other glasses such as fused silica may be used for UV-light applications. A polished, rugged metal housing, hermetically sealed to the window, guides photons to the photocathode and electrons to the first dynode. Focusing electrodes will be incorporated as needed. High-performance alkali-antimonide based photocathodes will be explored, and similar thin films will be considered for the secondary emitter to simplify fabrication. Figure 1.25 provides a simplistic diagram of the proposed design.

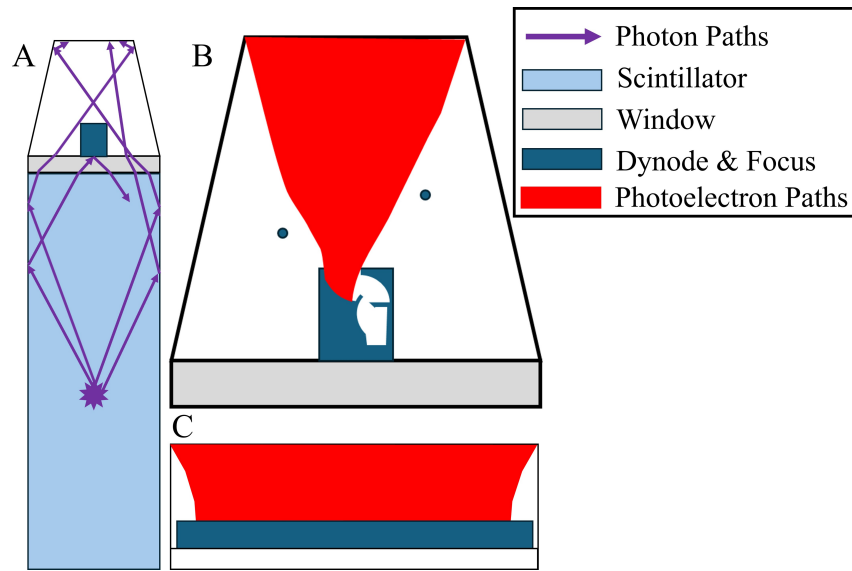


Figure 1.25: A) A cross-section of the reflective PMT concept mated to a rectangular scintillation crystal highlighting different photon paths to the PMT. B) A zoomed cross-section of the rectangular PMT concept illustrating photoelectron transit to the first miniaturized dynode stage guided by focus-wires, the PMT sidewalls, and the dynode potential. C) A side-profile of the reflective PMT showing the extended dynode stage and photoelectron transport.

Chapter 2

Scintillation Spectrometer Testing

Large-area PMTs can improve scintillation photon collection by covering the entire optical-coupling surface of large, rectangular scintillation crystals. However, photon collection is not the only metric to consider when designing a photomultiplier. Scintillation photons are produced with some spatial distribution within the crystal during broad-beam irradiation, meaning photoelectrons will also have some distribution across the photocathode dependent upon the gamma-ray interaction location and crystal geometry. Photoelectrons emitted over a large cathode area impinge the first dynode with a range of locations with trajectories that are highly contingent upon the initial emission location. Ultimately, this results in a spatially-dependent SER which increases the fractional variance of multi-electron signals. Spatially-dependent anode-response nonuniformity measurements were performed using a large-area PMT and a collimated photon source to provide a better understanding of how a PMT affects spectrometer energy resolution for systems composed of large crystals and photonic sensors. The large-area PMT was then coupled to scintillation crystals with drastically different geometries and optical interfaces. These crystals were irradiated in different locations by a collimated gamma-ray source to observe how spatial anode response nonuniformity for the coupled crystal/PMT system contributes to resolution for broad-beam irradiation.

2.1 PMT Characterization

A 130 mm diameter ET Enterprises 9490B PMT was acquired for spatial-nonuniformity testing. The 9490B has a reported 116 mm active diameter and uses a green-enhanced bialkali photocathode. 10 high-gain SbCs, linear-focus dynodes provide good timing characteristics and SER¹⁷. The wavelength-dependent QE was characterized by the manufacturer and can be seen in Fig. 2.1, below. A custom high voltage divider was also purchased from ET

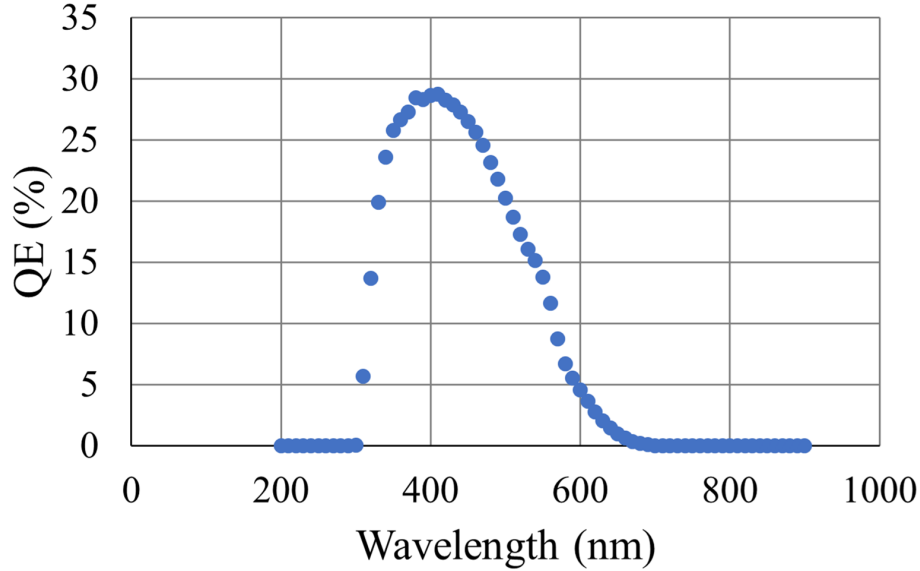


Figure 2.1: *Measured QE for the ET Enterprises 9490B PMT.*

Enterprises. The circuit diagram may be seen in Appendix C. Of note are the high-linearity tapered and transistorized (active) divider as well as the 5th-stage output, which may be used to test low-gain resolution performance such as that of the proposed device.

2.1.1 PMT Characterization Station

A large dark box (Fig. 2.2) was constructed to perform PMT characterization on the 9490B PMT. On the detector side, three BNC feedthroughs may be used to relay signals, while an MHV connector passes high voltage from a Caen DT8033 power supply to the PMT. An XY stage with built-in motion control from a modified laser-engraver is used to perform planar scans of the PMT photocathode. Custom brackets are used to mount collimated LEDs or

fiber-optic cables to the XY stage. LEDs provide a high intensity, narrow-band light source, while a Xe-plasma broad band light source paired to a monochromator may be used in the future for monochromatic studies of photocathode response across a range of energies.

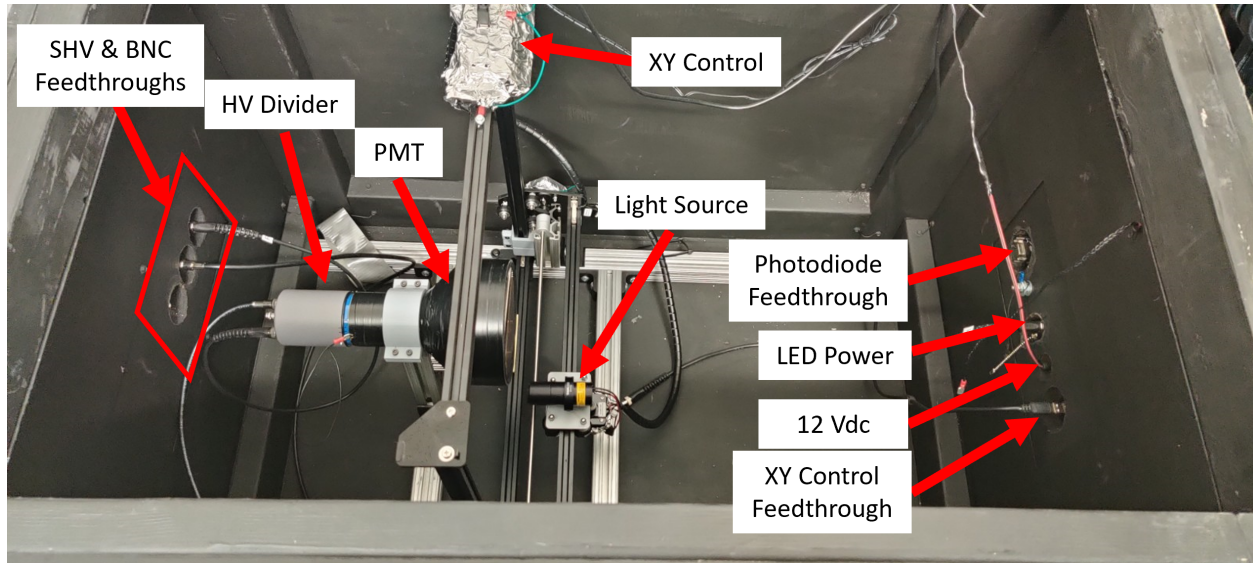


Figure 2.2: *Labeled components within the darkbox. Light traps and conductive shielding were incorporated to minimize noise.*

LEDs are powered by a Rigol DP831 DC power supply or a Sigilent SDG1020 function generator via BNC. An SMA-coupling feedthrough may be used to pass light from the plasma-based light source. A panel-mount DE-9 feedthrough is used to pass signal from the calibrated S120VC photodiode to the PM100USB power meter interface. A standard 5.5 mm coaxial DC connector supplies power from the DP831 to the XY-stage control and stepper-driver board. Directly powering the engraver using a DC power supply greatly reduced induced signal noise over the supplied 12 V AC-DC converter. A USB-A connector is used to pass commands from the computer to the control board. The connection diagram can be seen in Fig. [2.3](#)

Two custom scripts were written to pass g-code, operate the function generator, and correlate signal and positional information. The first script is used for constant-illumination, DC-signals as well as averaged current from pulsed signals through the Keithley 6487B picoammeter. Since the picoammeter is directly controlled through this script, position and signal information are directly linked. The alternative script may be used for pulsed-signals

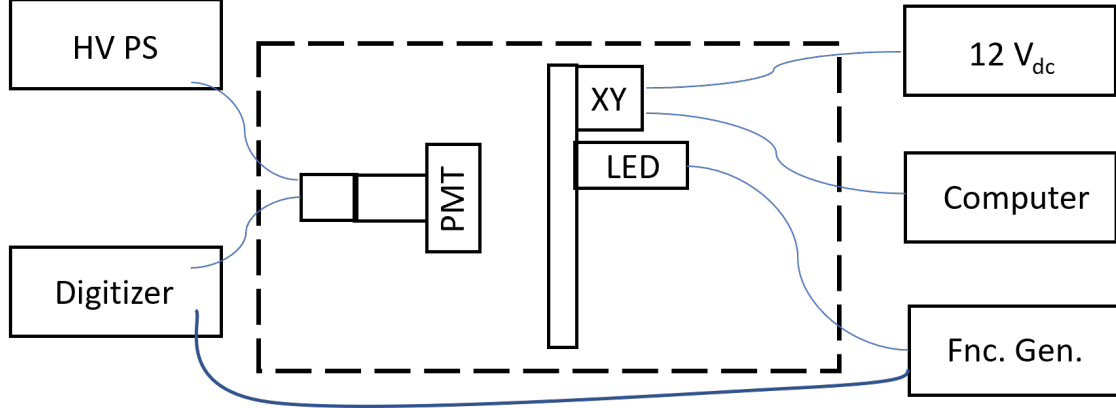


Figure 2.3: *The connection diagram for pulsed anode-response uniformity measurements.*

which are analyzed by a Caen DT5730 digitizer. Both channels on the SDG1020 function generator are used — one to power the LED, and the other to provide a signal gate. The PMT pulse-mode data and gate are recorded chronologically throughout a run. Another script is used to post-process the time-dependent integrated-charge distributions to produce a colormap of the mean anode response at each location. Both the mean anode response and deviation — which provides multi-photoelectron response characteristics such as the ENF — are recorded to better understand the contribution of response non-uniformity to spectrometer energy resolution. However, it is important to understand that the angular response is not captured through normal-incidence measurements such as these. Scintillation photons will have a largely-diffuse incidence on the PMT window. Depending on internal PMT structures, some of the collimated photons will preferentially reflect back to the photocathode at certain locations and angles of incidence, thereby increasing the PMT response in a manner dissimilar to that of a scintillator.

2.1.2 Anode Response Uniformity Measurement

Initial anode response uniformity measurements were carried out using the 9490B PMT and a Thorlabs LED545L LED. The LED545L is a 2.4 mW, 545 nm LED chosen to approximate the optical spectrum of CsI(Tl). The normalized emission distributions for both are shown in Fig. 2.4. The LED was assembled and housed using modular 25.4 mm diameter lens tubes

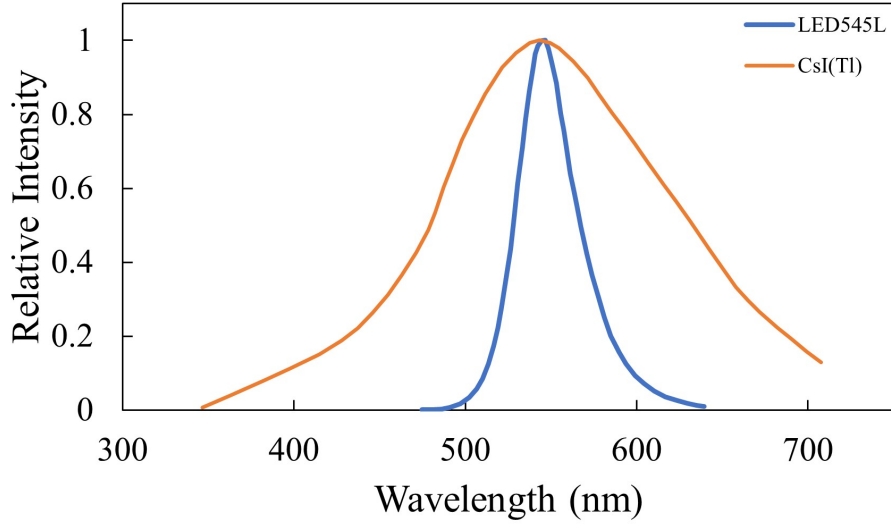


Figure 2.4: *The LED545L emission spectrum compared to a typical CsI(Tl) scintillator emission spectrum.*

and mounted orthogonally to the PMT window. Additional filters and pinhole collimators modify the beam intensity and diameter as illustrated in Fig. 2.5. The final beam diameter at the PMT window was calculated to be approximately 1 mm.

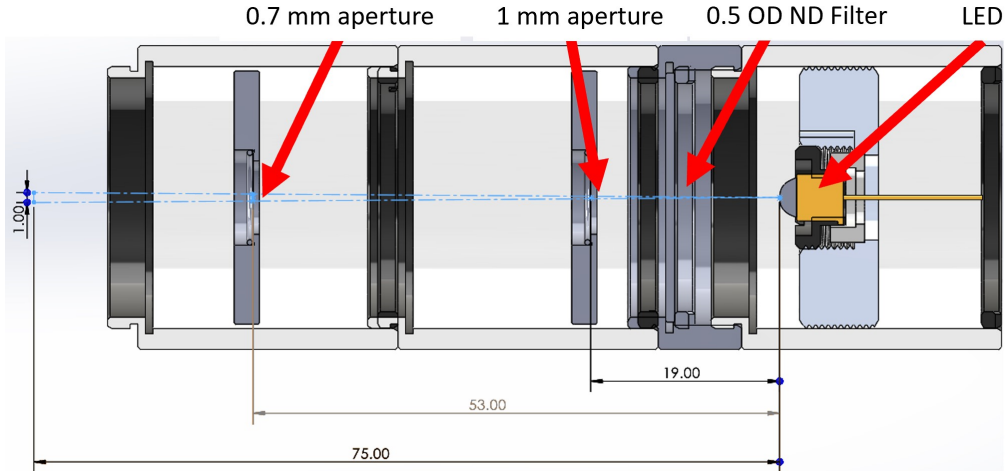


Figure 2.5: *The LED assembly used for 545 nm anode response measurements.*

Special care was taken to optimize the digitizer settings to ensure the gain and minimum charge threshold were appropriate for reliable measurements at the PMT periphery. The LED was fired with a trapezoidal, 200 ns (FWHM), 0-4.2 V pulse at 5 kHz. A 127 mm square grid was raster-scanned in 1 mm increments. 3-second measurements were taken at

each point with additional buffer time for motion and noise suppression. The integrated charge at the PMT anode was recorded for approximately 15,000 pulses at each location, and the results may be seen in Fig. 2.6. The total measurement time was 27 hrs.

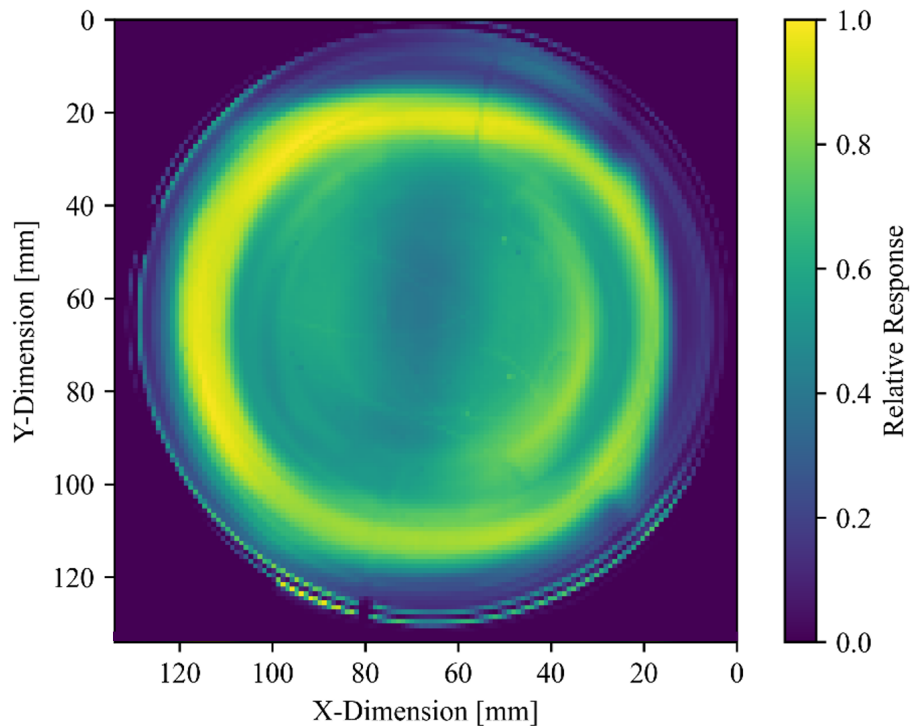


Figure 2.6: *The relative anode response heatmap for the 9490B PMT resulting from the 545 nm anode-response experiment.*

2.2 Gamma-Ray Spectrometer Response Characterization

Additional efforts are being undertaken to incorporate the PMT response into coupled gamma-ray/optical photon transport simulations with the goal of predicting the spectroscopic performance of novel systems prior to fabrication. To validate these simulations and better understand how PMT anode response nonuniformity affects spectrometer performance, collimated gamma-ray beam measurements were performed on two crystal geometries coupled the 9490B PMT. A sealed 150 mCi ^{137}Cs (300 mCi, assay date 5/31/1993) gamma

ray source was placed in a translating carrier behind 152.4 mm of lead. A 2 mm hole drilled through the lead shielding served as the collimator.

2.2.1 ^{137}Cs Beam Source Characterization

A $14 \times 14 \text{ mm}^2$, 256^2 pixel Advacam MiniPIX TPX was placed in the radiation path to image the beam profile. Multiple 10+ min. beam measurements were recorded at several distances from the lead-collimator face along the radiation path to characterize the beam divergence. Pixel binning was used as needed to reduce variance and improve beam profile analyses. A single profile measurement is provided in Fig. 2.7. The full-width at tenth-maximum

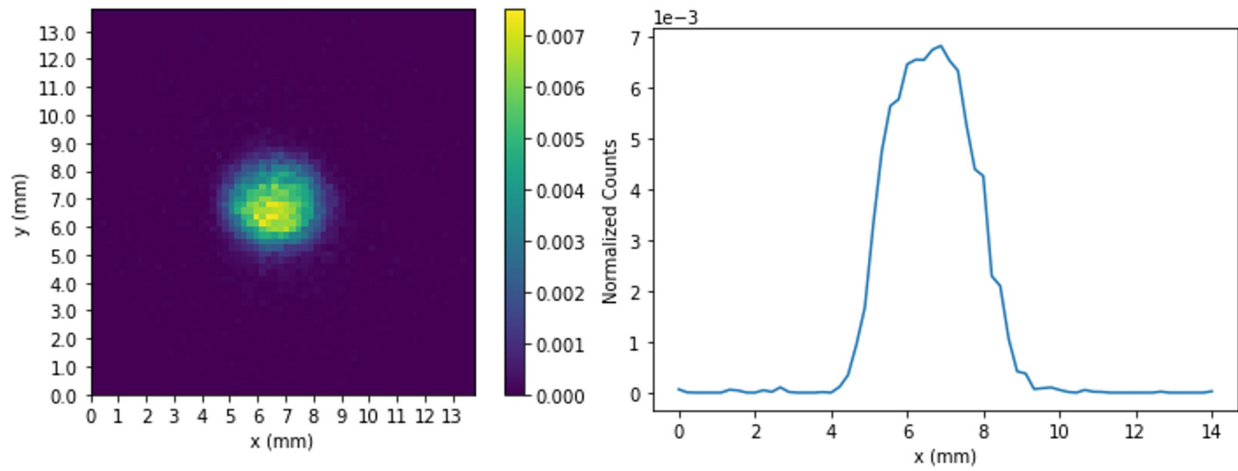


Figure 2.7: Left) The beam profile at 86 mm from the collimator assembly face. For the short 10 min. measurement, the data was rebinned to sum adjacent pixels and reduce noise. Right) A cross section of the collimated beam.

(FWTM) was calculated for each measurement location. The offset of the measured FWTMs from the 1 mm collimator radius are plotted in Fig. 2.8. The beam half-angle divergence was first calculated from a simple geometric approximation then compared to the experimentally derived half-angle divergence. Geometric approximation provides a half-angle divergence of $\theta_{est} = 0.72^\circ$, while the measured data reveals a half-angle divergence of $\theta_{meas} = 0.70^\circ$.

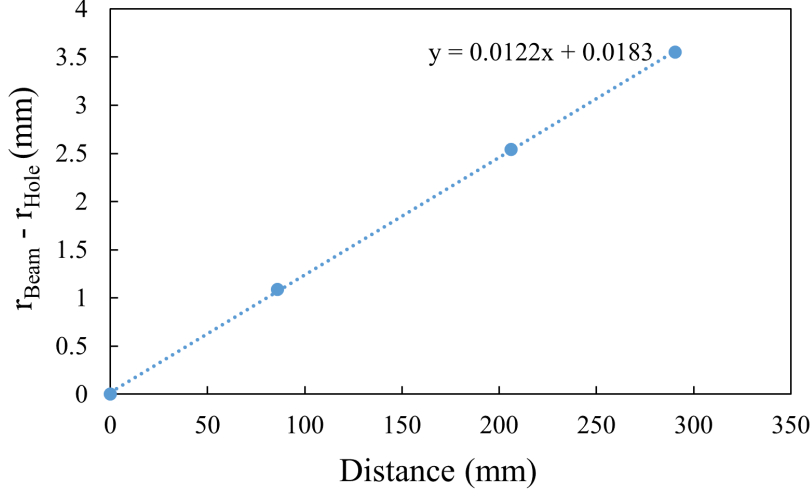


Figure 2.8: *The experimentally-measured offsets of FWTM from the 1 mm collimator radius for each test location shows how the gamma-ray beam spreads primarily due to collimator geometry. A linear regression of the beam diameter provides a half-angle divergence of $\theta_{\text{meas}} = 0.70^\circ$. The distance provided is measured from the lead collimator face.*

2.2.2 CsI(Tl) Scintillation Spectrometer Testing

Two CsI(Tl) scintillator crystals were tested using the beam source and a check source to quantify spatial uniformity and spectral resolution. Crystals with $25.4 \times 25.4 \text{ mm}^2$ and $25.4 \times 127 \text{ mm}^2$ optical interfaces were coupled to the 9490B window. Both crystals have the same outer dimensions, $25.4 \times 25.4 \times 127 \text{ mm}^3$. Therefore, the dimensions of the optical interface will be used first in the dimensional designation to identify each crystal. The $25.4 \times 127 \text{ mm}^2$ interface crystal was tested in two orientations to partially decouple the crystal positional response from the PMT response. Crystal mounting locations with respect to the relative anode response heatmap can be seen in Fig. 2.9.

Measurement Equipment The ET Enterprises 9490B PMT powered by the same Caen DT8033 power supply used for the PMT uniformity measurements. An ORTEC 113 preamplifier set to 1000 pF input capacitance was used to pass the signal to a Canberra 2022 shaping amplifier set at 8 μs shaping time for both CsI(Tl) crystals. The lowest achievable energy resolution was found by altering these parameters alongside the digitizer integration window. The amplifier gain was set to the minimum value of $3\times$ for all measurements, which

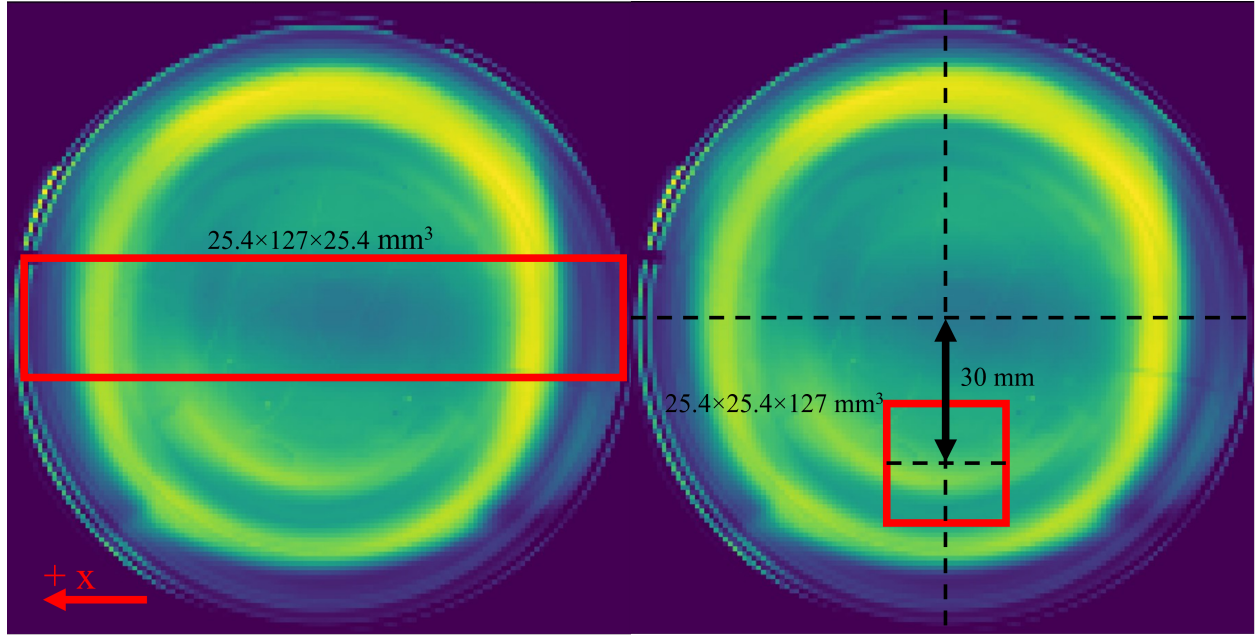


Figure 2.9: Red outlines overlaid on the 9490B PMT relative anode response heatmap indicate the two CsI(Tl) scintillators optical-coupling orientations. the $25.4 \times 127 \text{ mm}^2$ crystal was oriented in two directions, as indicated in Fig. 2.14

provided peak centroids ranging from channels 1350-1510 for 662 keV gamma rays fully absorbed by the $25.4 \times 127 \times 25.4 \text{ mm}^3$ crystal and channels 1420-1510 for the $25.4 \times 25.4 \times 127 \text{ mm}^3$ scintillator. A Tektronix TDS2004C was used for visual pulse analysis, while an Caen DT5730 digitizer provided pulse processing and spectral analysis capabilities. The equipment used to analyze spectrometer performance may be seen in Fig. 2.10.

The CoPASS software provided by CAEN was used to interface with the DT5730 digitizer, alter digitizer parameters, view waveforms, and collect spectra. A waveform produced by the $25.4 \times 25.4 \text{ mm}^2$ spectrometer and a ^{137}Cs check source may be seen in the digitizer and oscilloscope windows shown in Fig. 2.11. The optimal pulse-integration window was $32.7 \text{ } \mu\text{s}$, which is similar to that found by Mianowska for CsI(Tl) using a $10 \text{ } \mu\text{s}$ shaping time⁷⁰.

Initial ^{137}Cs γ -ray beam testing of both CsI(Tl) crystals revealed a consistent, monotonic $1.1 \pm 0.2\%$ increase in the peak centroid channel over time. This effect is ascribed to increased scintillation light output as a result of deep level trap filling and is known as “sensitization”, “bright burn”, or “hysteresis”. Once filled, these traps can no longer accept free

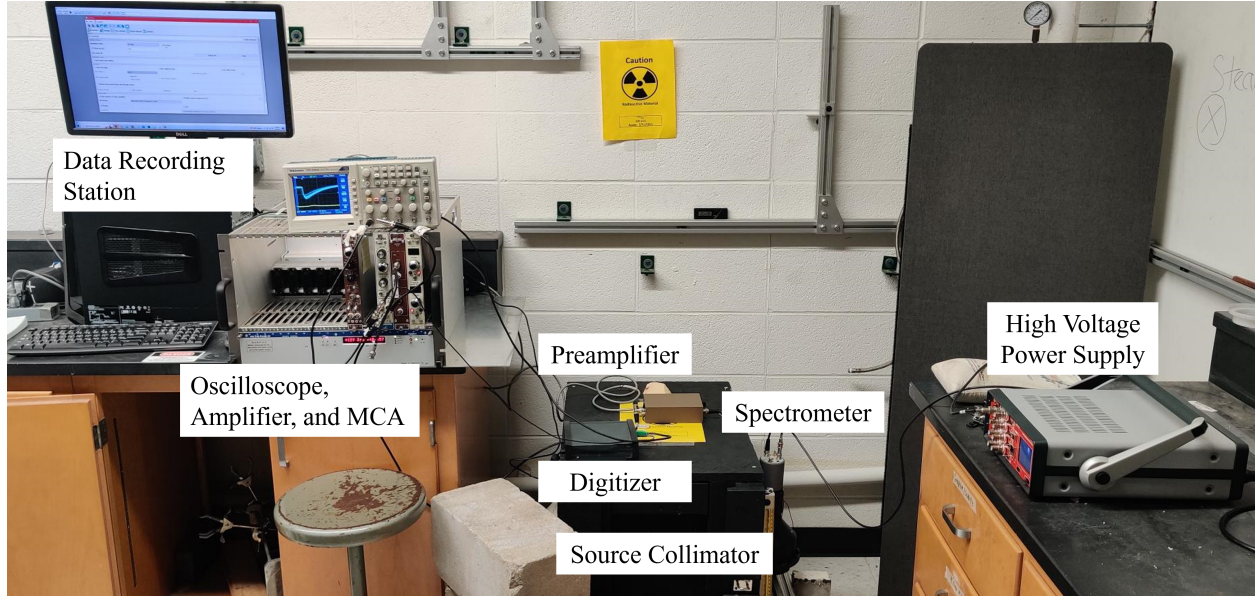


Figure 2.10: The experimental setup for spectrometer-uniformity measurements using the collimated ^{137}Cs source.

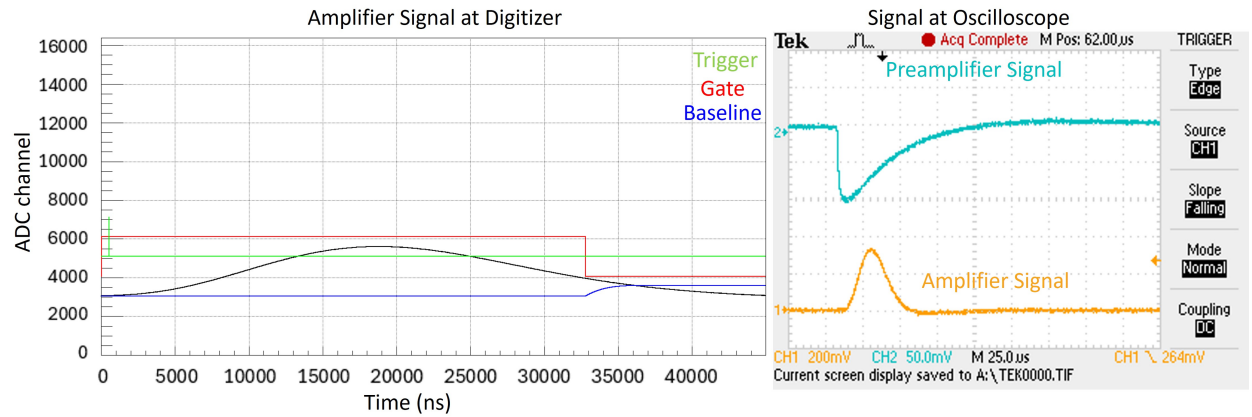


Figure 2.11: Signal pulses from the amplifier and preamplifier produced by the $25.4 \times 25.4 \text{ mm}^2$ CsI(Tl) spectrometer. A long $32.7 \mu\text{s}$ integration window provided the lowest energy resolution for the relatively slow CsI(Tl) spectrometer.

carriers, thereby increasing the probability for charge carriers to fill luminescent centers^{4;71}. This positive sensitization is not attributable to radiation damage, which typically reduces light output by enhancing self-attenuation, nor can it be ascribed to temperature-related effects (increased yield or improved emission/QE spectral matching) in the climate-controlled laboratory. Further testing incorporated a tail-pulse generator as a spectrometer-surrogate to inject full-energy-peak-equivalent pulses at frequencies up to $4\times$ that of the total γ -ray

count rate, ultimately concluding that it was not a rate-induced effect. Another “memory effect”, the well-characterized afterglow of CsI(Tl)^{4;70–72} was ruled out as well, since the slow phosphorescence will not contribute meaningfully to a capacitively-coupled HV-divider. For example, the CsI(Tl) crystals grown and characterized by Tosuka⁷² exhibited a multicomponent decay, with significant contributions having time constants that range from $10^{-3} - 10^6$ s.

The sensitization rate and magnitude are dependent on material properties, source activity, and temperature⁷¹. Nine sensitization measurements were taken between the two crystals with the ^{137}Cs beam source continually irradiating the center of the scintillators. The results were fit using the equation $S(t) = S_0 + A(1 - \exp(-t/\tau_s))$, where S is the full-energy peak centroid channel, S_0 is the full-energy peak centroid channel at time $t = 0$, A is a scaling parameter representing the increase in full-energy peak centroid channel, and τ_s is the sensitization time constant. The fits were then normalized by setting $S_0 = 0$ and $A = 1$, after which the mean and standard deviation of the sensitization time constants were calculated. For the ^{137}Cs beam source impinging both CsI(Tl) crystals, $\tau_s = 310 \pm 50$ s. Figure 2.12 shows the normalized sensitization curve modeled using the average and standard deviation of the fits as well as the raw data and individual fits.

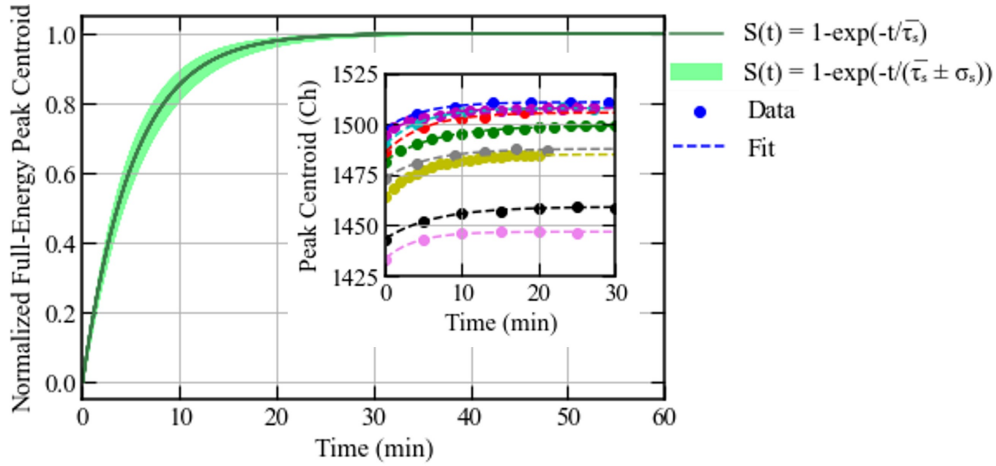


Figure 2.12: The normalized sensitization fit, derived from the mean and standard deviation of individual sensitization fits. Inset) The raw sensitization measurement data and individual fits used to calculate the sensitization rate. This data led to the implementation of a 30-min. irradiation period prior to spatial spectrometer nonuniformity measurements.

The sensitization decay rate was also measured for the two scintillators. First, the beam source was used to sensitize the crystal over a period of approximately 30 min. Once the peak centroid had plateaued at the upper limit, the beam source was shut off, and a series of two-minute check source measurements were taken. The check source was used since its comparatively low activity had no discernible effect on the scintillation yield. The “desensitization” data was fit using the exponential decay equation, $S(t) = S_0 + A\exp(-t/\tau_d)$, where τ_d represents the desensitization time constant. The measured data was again fit, then normalized (Fig. 2.13). For the $25.4 \times 25.4 \times 127$ mm³ CsI(Tl) scintillators, the desensitization time constant was $\tau_d = 1460 \pm 380$ s. The small 15-20 channel difference in the

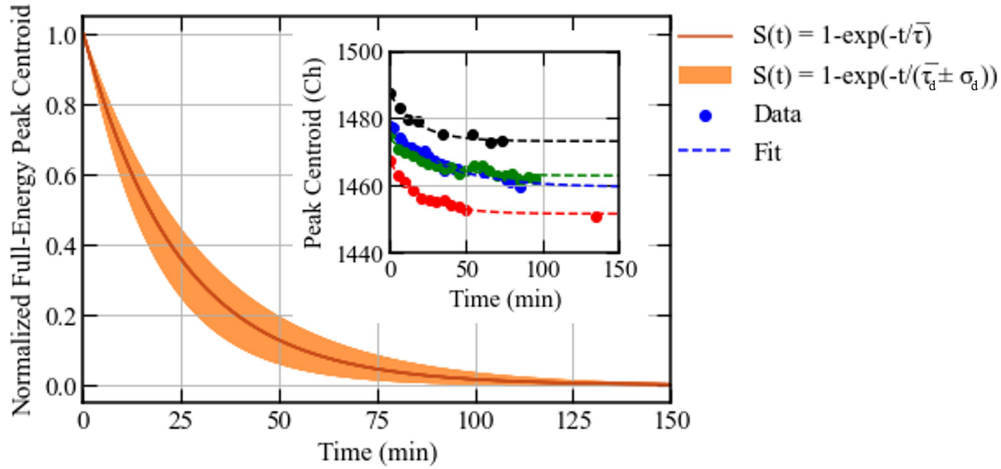


Figure 2.13: *The normalized desensitization fit, derived from the mean and standard deviation of individual decay fits. Inset) The raw desensitization measurement data and individual fits used to calculate the decay rate. The comparatively slow decay rate meant that spatial nonuniformity runs could be performed without significant pulse-magnitude reduction.*

peak centroid, though observable in the differential pulse-height spectrum, had no effect on the energy resolution outside of the uncertainty of the peak fit, which was primarily dictated by counting statistics. For the beam-source sensitization measurements, the standard deviation of the resolution measurements was $\sigma_{res} = 0.3\%$ of the mean, whereas the check source desensitization measurements had a $\sigma_{res} = 1.6\%$ of the mean due to the lower number of counts in the full-energy peak.

25.4×127×25.4 mm³ CsI(Tl) Scintillator The 25.4×127×25.4 mm³ CsI(Tl) scintillator — roughened, wrapped in a diffuse-reflector paper, and backed by a thin reflective tape on each surface except the optical boundary — was tested multiple times in two, opposing orientations, as shown in Fig. 2.14. Nine ¹³⁷Cs beam measurements, having a diameter of

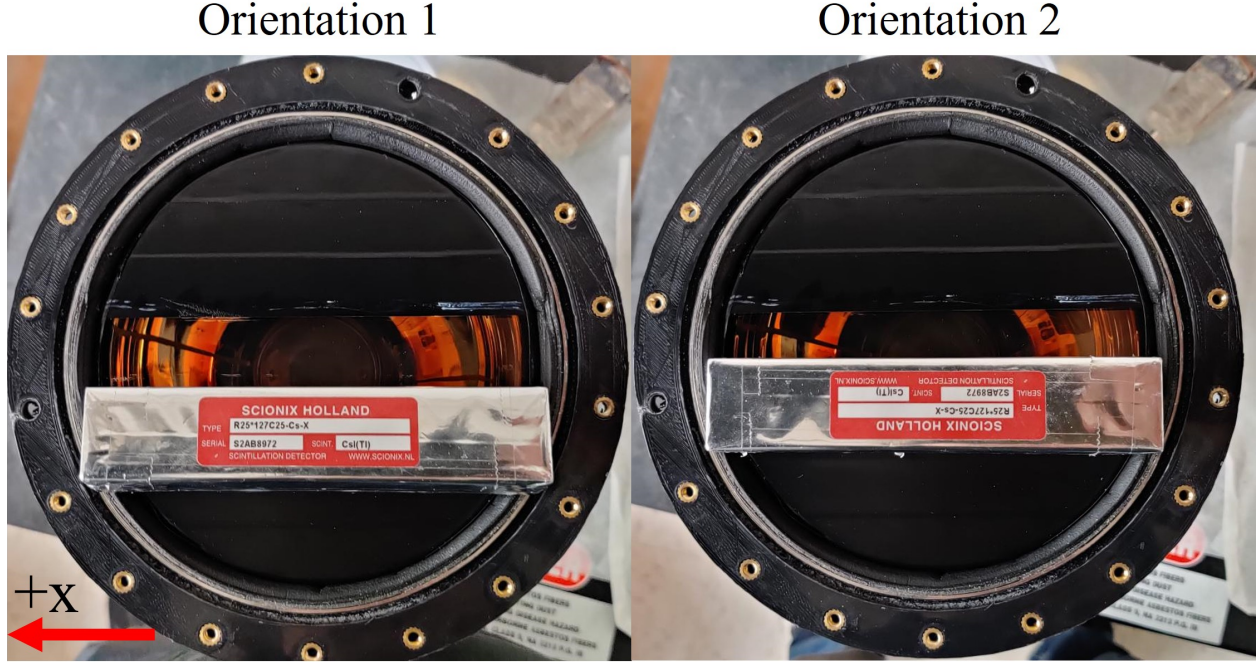


Figure 2.14: *Orientation of the 25.4×127 mm² CsI(Tl) crystal on the PMT for the two experiments.*

4.7 mm at the scintillator’s center plane, were taken for each run. A diagram of the source geometry and beam-incidence locations may be seen in Fig. 2.15. A series of 2 min. 0.87 μ Ci ¹³⁷Cs (1 μ Ci assay date Feb. 2018) check source measurements and 1 min. beam-source measurements were taken prior sensitization to determine a baseline full-energy peak centroid and resolution. A 30 min. center-irradiation was performed before each run to sensitize the crystal prior to spatial measurements. Six total runs were performed; three runs per orientation with varying measurement order to decouple (de)sensitization from the measurement time and location. Once an optimal crystal-PMT coupling was confirmed through measurement, the spectrometer was left assembled until all three runs were performed for the given orientation to ensure consistency between runs.

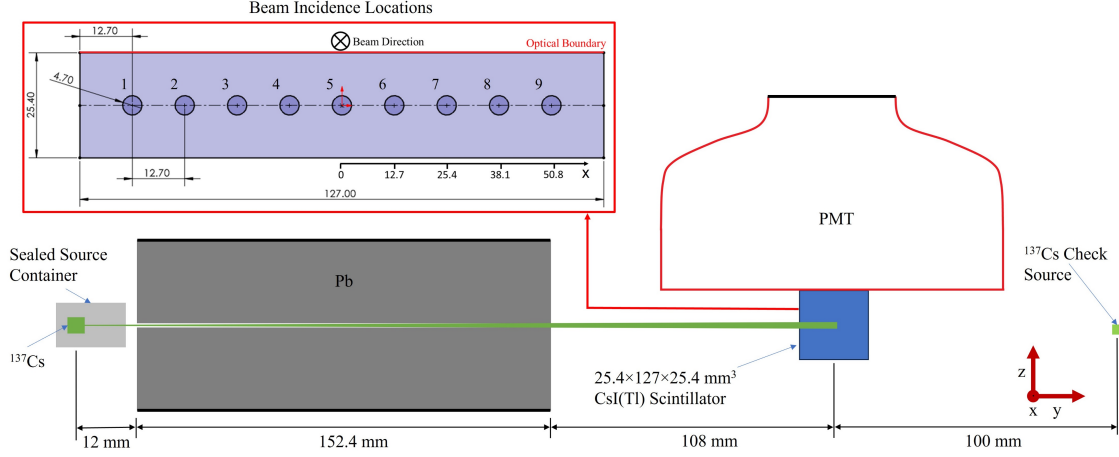


Figure 2.15: A diagram of the ^{137}Cs collimator and $25.4 \times 127 \text{ mm}^2$ CsI(Tl) spectrometer assemblies illustrates the measurement layout. An inset diagram (top left) of the $25.4 \times 127 \text{ mm}^2$ CsI(Tl) crystal cross section shows the beam incidence locations and beam diameter at the crystal's center plane.

Analyses of the resultant spectra were performed using CoMPASS. A simple linear fit was deemed sufficient to subtract background from the full-energy peak. A ^{137}Cs check source spectrum for the $25.4 \times 127 \text{ mm}^2$ CsI(Tl) scintillator may be seen in Fig. 2.16. Each

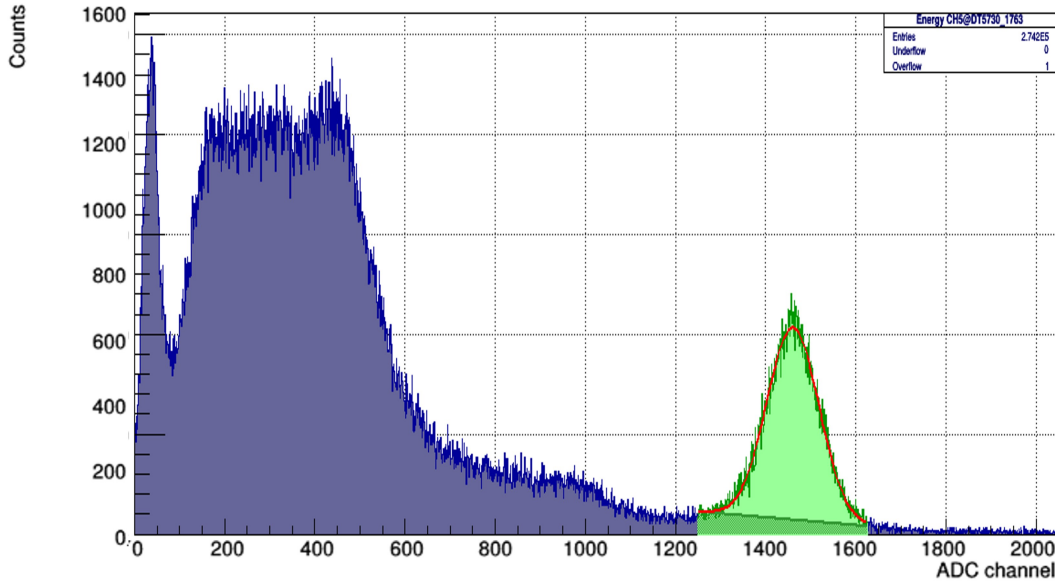


Figure 2.16: A ^{137}Cs check source spectrum for the $25.4 \times 127 \text{ mm}^2$ CsI(Tl) scintillator plotted in CoMPASS shows the Gaussian full-energy peak fit (red line), the gross full-energy peak counts (green shaded area), the linear background fit (green line), and the net full-energy peak counts (light green shaded area). The large number of background counts are the result of downscatter leakage from the 150 mCi ^{137}Cs collimator assembly.

measurement taken during the three runs (per orientation) were normalized to the mean check source full-energy peak centroid taken after positional measurements. Normalized values for each run were then averaged at individual irradiation-locations. Figure 2.17 shows the averaged, normalized spatially-dependent full-energy peak centroid and average energy-resolution data for each location along the x -axis. The short, wide $25.4 \times 127 \times 25.4 \text{ mm}^3$

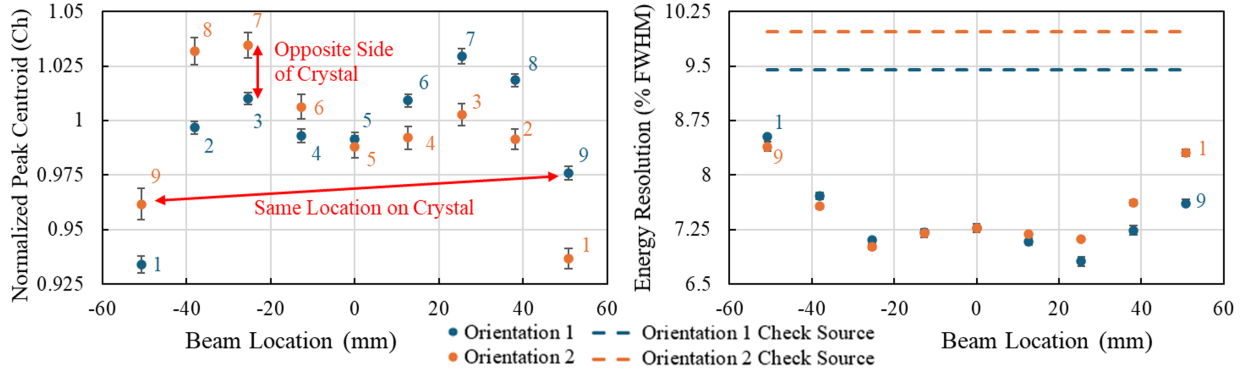


Figure 2.17: Left) Average normalized ^{137}Cs beam-measurement full-energy peak centroids for each measurement location and orientation. Right) The average spectrometer energy resolution for each beam measurement plotted against the broad-beam check source energy resolution for each measurement location and orientation.

scintillator produces a unique position-dependent response. Because scintillation photons are produced and reflected in close proximity to the photocathode, the signal appears to be dependent on the spatial PMT anode response. Looking at the relative PMT anode response

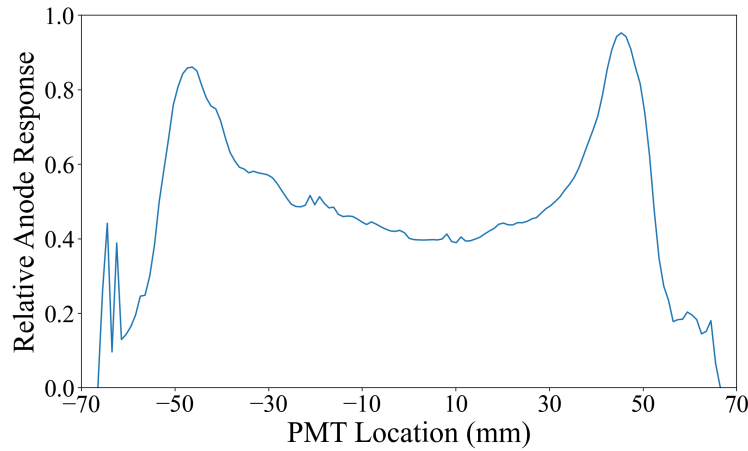


Figure 2.18: The cross section of the measured 9490B relative anode response centered on the $25.4 \times 127 \text{ mm}^2$ CsI(Tl) optical boundary.

along the x -axis in Fig. 2.18, one can see that the highest spectrometer responses occur away from the sidewalls, near the peak relative anode response surrounding ± 25.4 and ± 38.1 mm. Conversely, one may also observe lower full-energy peak centroids at 0 mm and ± 50.8 mm potentially caused by lower PMT anode response. Light collection efficiency likely suffers from increased boundary-absorptions at ± 50.8 mm⁷³.

Relatively short, 2-3 min. check-source measurements were also taken before and after each run to analyze the scintillator response to broad-beam irradiation. Short measurements were required to avoid significant desensitization over the measurement period. As expected, the variable response across the scintillator seen in ^{137}Cs beam-source measurements produces a larger spread in energy resolution, as shown in Fig. 2.17. The background-subtracted beam-measurement and check source spectra from a single run (for each crystal orientation) were normalized to one. The beam-measurement spectra were further scaled by a geometric weighting factor to account for isotropic emission from the check source 100 mm away from the detector's mid-plane (Fig. 2.15). The geometric weighting factors were calculated as the angle subtended by the width of individual volumes — each corresponding to one of the nine beam-measurement locations. These values were then normalized such that the sum of the weighting factors was equal to one. The results of these analyses are shown in Fig. 2.19. The sum-peak fits quite well for both orientations, revealing that the broad-beam spectra can indeed be modeled as a summation of spatially-dependent responses.

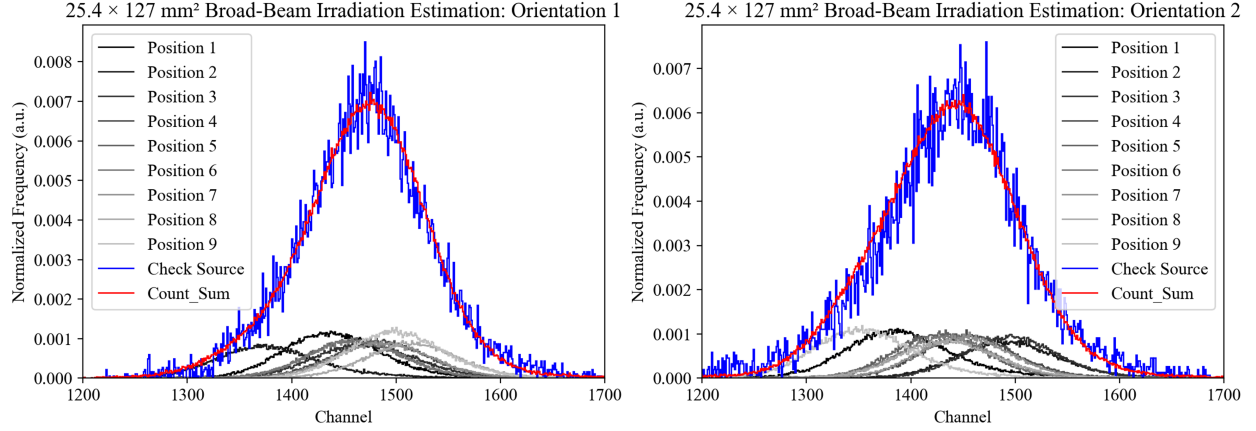


Figure 2.19: Left) Full-energy peak spectra for Orientation 1. Right) Full-energy peak spectra for Orientation 2. The individual, scaled ^{137}Cs beam-measurement spectra (Positions 1-9) are plotted in grayscale. The summations of the scaled beam-spectra are provided in red, and the check source spectra are shown in blue. Check source measurements are noisier as they contain fewer full-energy peak counts.

25.4×25.4×127 mm³ CsI(Tl) Scintillator Figure 2.20 illustrates the source-detector orientation and the beam measurement locations for the 25.4×25.4×127 mm³ CsI(Tl) crystal. The scintillator has polished sidewalls, and is packaged in a diffuse-reflector paper backed by a specular-reflector tape.

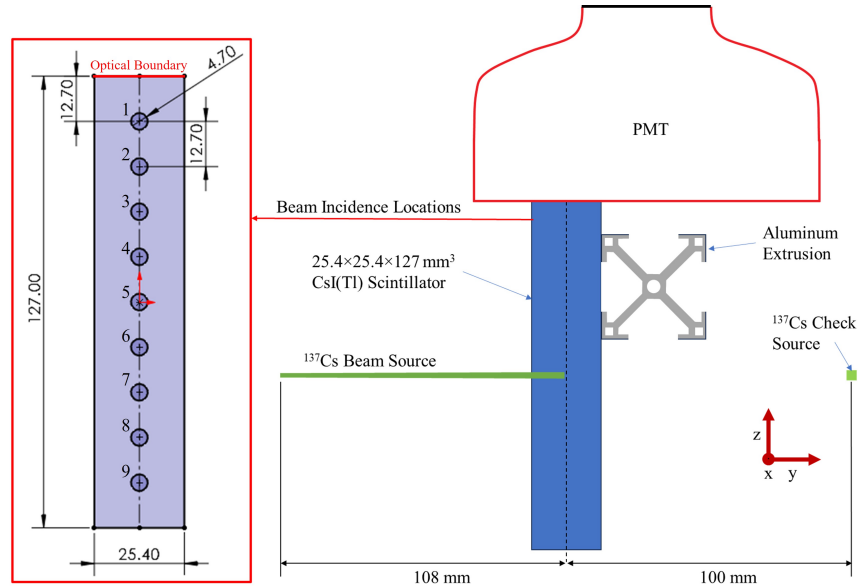


Figure 2.20: A diagram of the ^{137}Cs beam source and 25.4×25.4 mm² CsI(Tl) spectrometer assembly. An inset diagram (left) of the 25.4×25.4 mm² CsI(Tl) crystal cross section shows the beam incidence locations and beam diameter at the crystal's center plane.

The primary goal of these tests was not to validate the incorporation of PMT nonuniformity in coupled gamma-optical simulations, but to provide a validation model for the behavior of simulated high aspect ratio crystals. For these measurements, the crystal was not reoriented between runs, since the optical interface is relatively small. Three runs at $x = 0$ mm were performed with the same coupling to ensure consistency. Two runs at $x = \pm 6.35$ mm were also performed, with these "off-center" measurements as a way to observe edge-effects caused by interactions near the reflective sidewalls. Figure 2.21 shows the PMT and crystal interface.

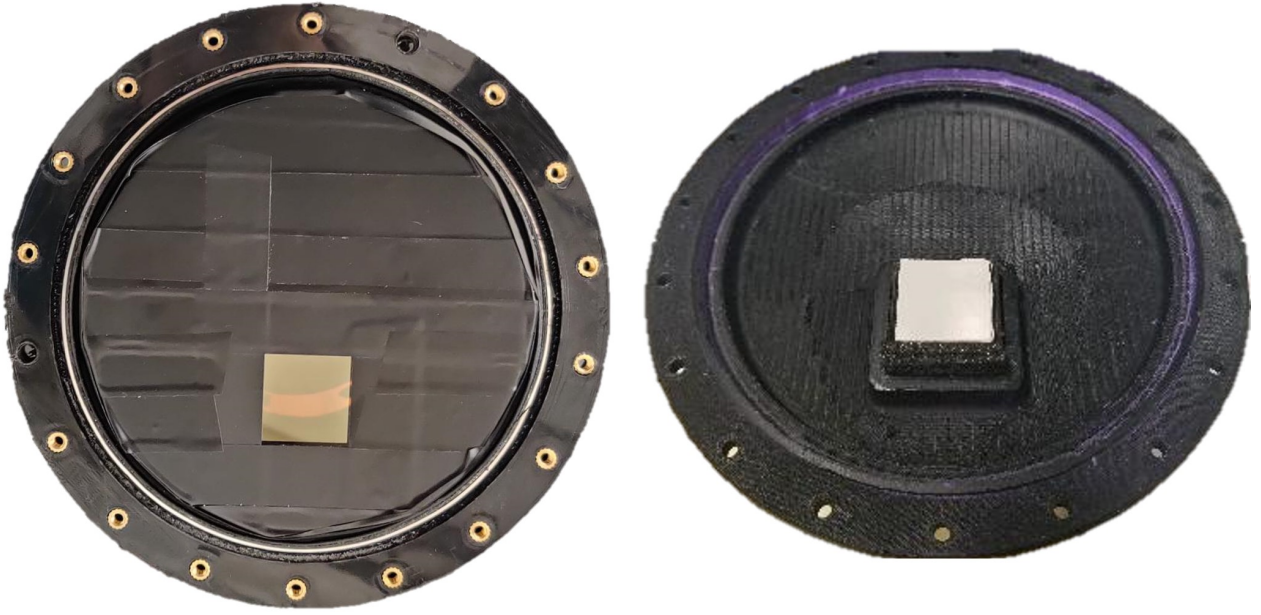


Figure 2.21: *Left) The PMT window with black electrical tape applied to reduce extraneous reflections. Right) The 25.4×25.4 mm² CsI(Tl) surface contained within the crystal-side of the spectrometer housing.*

An example beam spectrum, captured in CoMPASS, is shown in Fig. 2.22. Compared to the check source measurements, the beam measurements contain relatively little scatter, and far more full-energy counts.

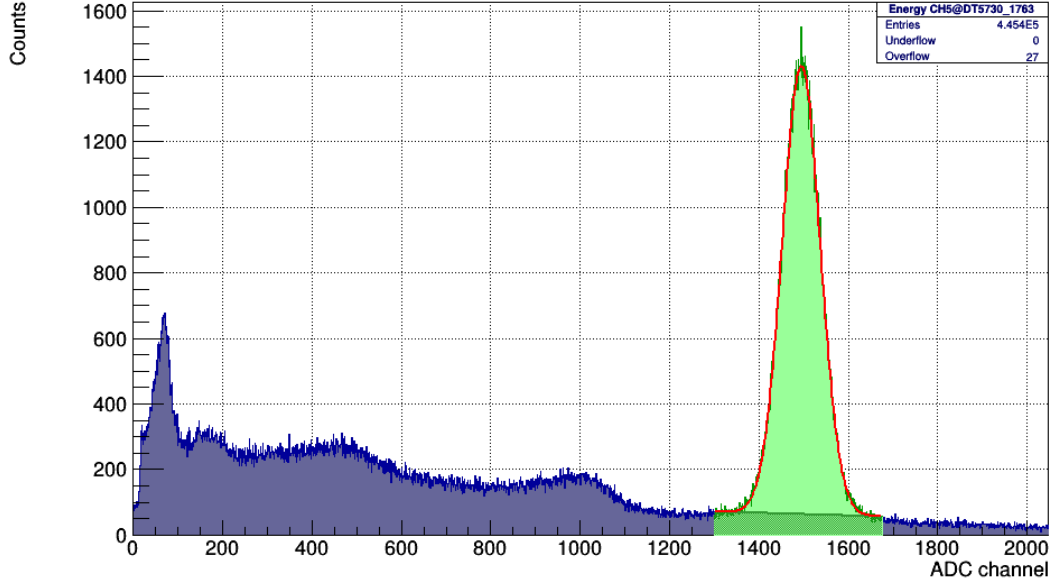


Figure 2.22: A ^{137}Cs beam-source spectrum taken with the $25.4 \times 25.4 \text{ mm}^2$ CsI(Tl) scintillator. Again, a linear fit was used to subtract the background underlying a Gaussian fit to the full-energy peak.

Similarly to the $25.4 \times 127 \text{ mm}^2$ scintillator, the full-energy peak centroid at each measurement location was normalized to that of the check source measurement for each run. The energy resolutions are also plotted for comparison against the check source in Fig. 2.23. Scintillations occurring near the PMT window had a markedly lower response, due to poor

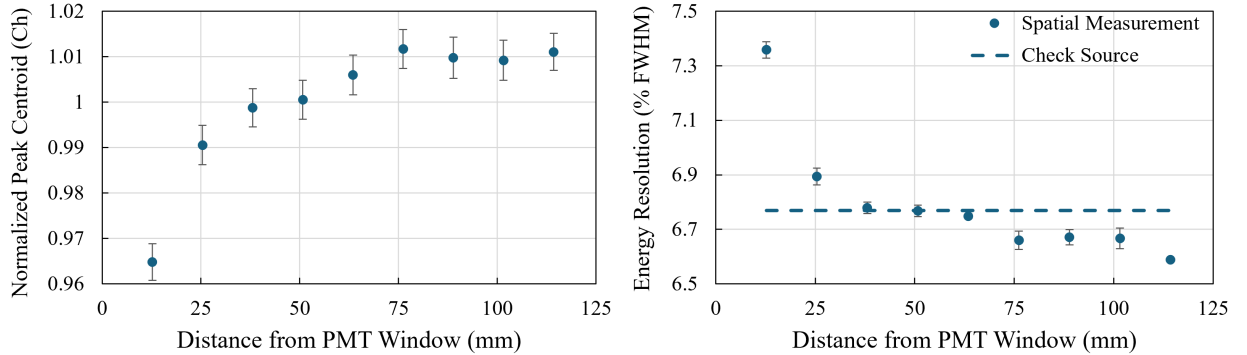


Figure 2.23: Left) Average normalized full-energy peak centroids for several ^{137}Cs beam-measurements taken at various distances from the optical coupling. Right) The resolutions of each measurement location were averaged and are compared to the averaged broad-irradiation check source measurements.

light collection efficiency. The light collection efficiency and energy resolution appear more

uniform towards the bottom of the crystal, and the fractional variance of all measurements is much lower than that of the $25.4 \times 127 \text{ mm}^2$ scintillator, meaning that the detector resolution is much lower as well — even though the maximum peak centroid channel, and therefore integrated charge, is similar for both scintillators at approximately 1500.

The nine individual beam measurements were normalized, scaled by the geometric weighting factor, summed, and compared to the check source measurement (Fig. 2.24). The initial comparison revealed excess counts in the low-end of the full-energy peak attributable to Position 1 (closest to the PMT). Upon further scrutiny, it was realized that a significant obstruction was attenuating check-source emissions near the optical interface. Figure 2.20 shows the obstruction: a $38 \times 38 \text{ mm}^2$ aluminum t-slot extrusion. Subsequent analysis omitting the Position 1 measurement provides a much better estimation of the check source spectrum; however, a true representation of the attenuation would likely produce a sum-spectrum somewhere between the two provided.

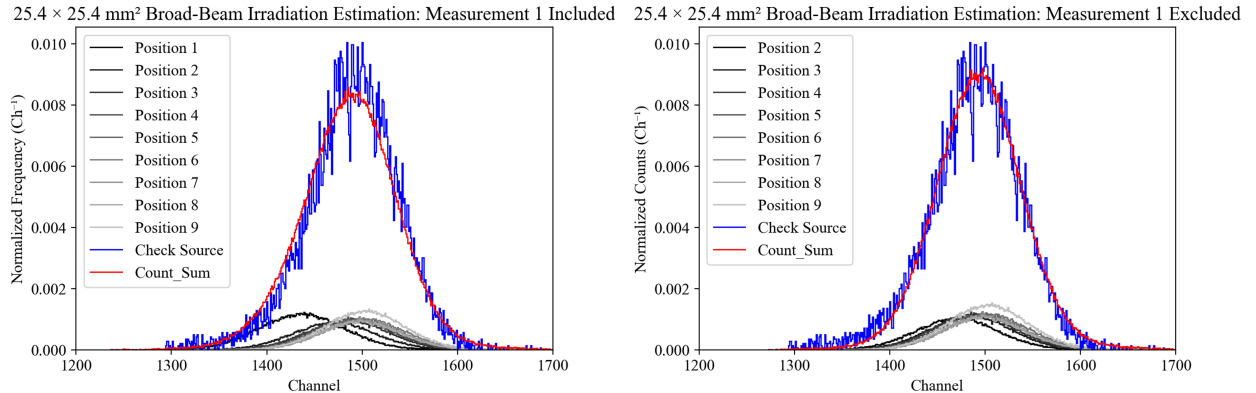


Figure 2.24: *Left) The full-energy peak spectra for several positional measurements and the check source. Right) The same spectra with the Position 1 data omitted due to an obstruction between the check source and scintillator. Again, the sum-peak reconstructs the broad-beam check source spectrum quite well.*

Positional beam-source measurements were again performed; this time for both centerline and off-center measurements. Figure 2.25 shows the full-energy peak centroid and resolution data for the 15 measurements. Although the full-energy peak centroids and resolutions are nearly equivalent at 38.1 mm away from the PMT window and beyond, the data is quite disparate at 12.7 mm, indicating a spatial dependence in light collection efficiency.

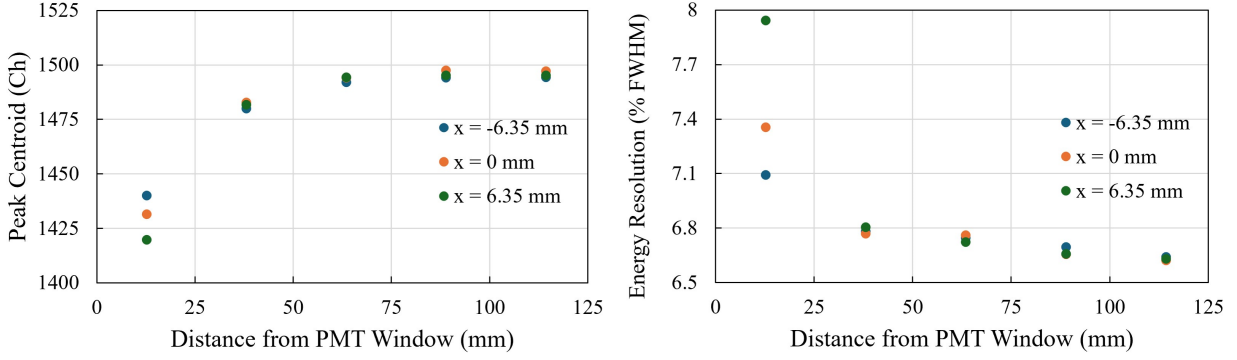


Figure 2.25: *Left) Full-energy peak centroids taken throughout the CsI(Tl) crystal reveal a difference in response across the crystal width near the PMT window. Right) Similarly, the different energy resolutions at $z = 12.7$ mm shows the effects of reduced photoelectron emission. The close proximity of each measurement at $z = 12.7$ mm suggests variability in photon collection as opposed to photon generation.*

2.3 Conclusions

Several studies were performed to explore the spatial anode-response uniformity of large-area PMTs and its effect on gamma-ray spectrometer performance pertinent to the development of a novel rectangular PMT. Though not exhaustive with regards to spectral-dependence, angle of incidence, and scintillator geometry, the testing provided insight and infrastructure needed for continued testing and coupled gamma-ray and optical photon transport simulations in Geant4. A number of conclusions may be drawn from the results of the PMT and spectrometer testing.

2.3.1 PMT Anode Response Uniformity

A 1 mm diameter, 545 nm LED was scanned across the photocathode of a 127 mm, green-enhanced ET Enterprises 9490B PMT. The anode-response heatmap shows a large variation across the PMT face, and the cross section provided in Fig. 2.18 reveals a factor of two difference between the maximum and minimum anode response over the sensitive area. Such photocathode nonuniformity is common, even in smaller 50 mm PMTs⁷³, and it is often attributed to back-reflections and dynode response nonuniformity rather than cathode film nonuniformity¹⁸. For orthogonal beams, especially, back-reflection may locally enhance the

anode response by effectively increasing the QE, since photons have a second opportunity to produce photoelectrons. The angled, metalized sidewall of the 9490B PMT focuses photoelectrons towards the first dynode, but photons may also be redirected towards the photocathode as shown in Fig. 2.26. Future studies may vary the angle of incidence to decouple the anode

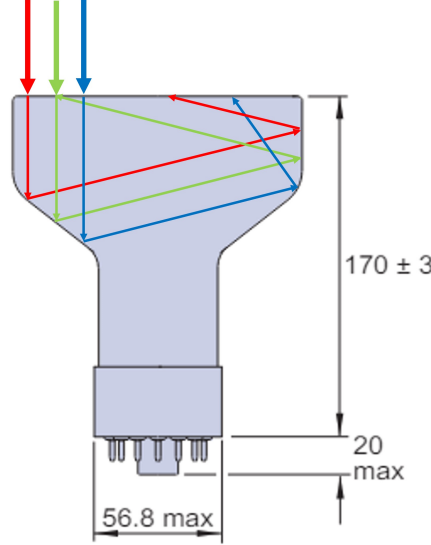


Figure 2.26: Red, green, and blue arrows indicate potential optical paths as a result of transmission and back-reflection within the PMT. 9490B PMT cross-section image provided by ET Enterprises¹⁷. Dimensions are in mm.

response from the incidence location. A diffuser may also be incorporated to emulate the diffuse photon emission of scintillators. Studies at various wavelengths are still needed to map the PMT anode response for other scintillators such as NaI(Tl) and LaBr₃(Ce). QE and single-photoelectron studies are also needed to estimate the multiplier resolution, R_m . Ultimately, the spatially-dependent relative anode-response is needed to accurately predict the resolution of novel PMT and crystal geometries via simulation.

2.3.2 CsI(Tl) Scintillation Spectrometers

Irradiating both CsI(Tl) crystals with the ¹³⁷Cs beam-source revealed a sensitization effect, wherein the scintillation light yield increased over time dependent upon the gamma-ray source characteristics. While relatively small, with a steady-state full-energy peak centroid 1-2% above the baseline, the effect makes a noticeable difference in precision mea-

measurements. The energy resolution is largely unaffected by short duration measurements as the full-energy peak centroid is consistent over the measurement period, although longer beam-source measurements exhibit higher energy resolution due to centroid-shifting. Sensitization was performed prior to spatially-dependent measurements to mitigate full-energy peak centroid shifting. Additionally, the measurement order was varied to decouple time-dependence. Differences between identical runs are primarily attributed to small changes in the ADC calibration of the DT5730 digitizer. Full-energy peak data normalization was used to effectively circumvent minor changes in the calibration between runs.

2.3.3 $25.4 \times 127 \text{ mm}^2$ CsI(Tl) Scintillator

The $25.4 \times 127 \text{ mm}^2$ CsI(Tl) crystal exhibited a large variance in full-energy peak centroid for ^{137}Cs beam-source measurements resulting in a relatively high energy resolution. Testing the crystal in two opposing orientations produced unique spatially-dependent responses, with the results being approximate reflections of each other. This result implies that the crystal or packaging is the source of the disparity. Additional coupling/decoupling experiments support this conclusion, since the result is repeatable. Small differences between results when mirroring one orientation may be attributed to the PMT anode response variation; however, the differences mostly lie within the standard deviation of the normalized data. Although the “saddle-shaped” trend of the the full-energy peak centroids may be a consequence of the crystal geometry and optical transport, the shape is reminiscent of the PMT anode response cross-section shown in Fig. 2.18. The difference between response extrema is comparatively muted in the full-energy peak centroid data, but this may be explained by the scintillation and reflected-photon distributions. Coupled gamma-ray and optical photon transport simulations may help elucidate the source of this trend. Similarly, experiments using the same CsI(Tl) crystal, segmented into smaller volumes, would reveal any light output nonuniformity inherent to material differences. Variable surface-roughness and reflector quality or optical grease ingress may also contribute to disparate light collection ^{18;73;74} and may be resolved by resurfacing and rewrapping the crystal. Strategic modification of the

crystal surface and/or reflector properties could be used to match localized scintillator light output to that of the PMT anode response. However, the asymmetric PMT anode response necessitates similarly asymmetric alterations to the scintillator and/or packaging.

2.3.4 $25.4 \times 25.4 \text{ mm}^2$ CsI(Tl) Scintillator

Positional beam-source experiments of the $25.4 \times 25.4 \times 127 \text{ mm}^3$ CsI(Tl) crystal revealed a much smaller variance in full-energy peak centroid and lower energy resolution compared to the $25.4 \times 127 \times 25.4 \text{ mm}^3$ CsI(Tl) crystal, even though the total light output was approximately the same. Through these measurements, it is clear that the crystal response is much more uniform along its length. Measurements at several locations along the x -axis show a spatial dependence on light collection efficiency near the PMT window partly attributed to grease ingress⁷⁴, which was visible after removing the crystal from the PMT window. The resultant reduction in reflectivity may be the source of lower light-output near the PMT. Additionally, photons emitted near the PMT window with trajectories having a $-z$ component must travel much further and undergo many more boundary interactions enhancing self- and boundary-absorptions compared to those produced further away from the PMT.

Uniform light output along the crystal length, beyond 50.8 mm suggests a quality diffuse reflector⁷³, as photons emitted near the crystal base are not subjected to higher boundary absorption. The diffuse-reflector wrapping promotes wide-angle reflections which reduce the probability of photon trapping due to purely specular reflection. Furthermore, by using polished sidewalls for the high aspect ratio crystal, shallow incidence angle photons undergo total internal reflection as they travel to the photocathode.

2.3.5 Relevance to the Proposed Rectangular PMT

Oftentimes, commercial spectrometer assemblies will incorporate a relatively thick window or light-guide to separate the photocathode from the crystal to reduce the effect anode response nonuniformity has on thin-crystal resolution^{18;73}. This separation more widely distributes scintillation light over the photocathode and decouples the PMT anode response

nonuniformity from the scintillation location. Thin crystals with large optical interfaces, especially, suffer from photocathode nonuniformity, since the light is highly localized on the photocathode through both direct transmission and immediate back-reflection⁷³. This effect is further exacerbated for charged particles and low-energy gamma rays, which deposit energy in a relatively small volume. Offsetting the photocathode from the optical window, as in the proposed reflective photocathode PMT, provides another method of widely distributing scintillation photons. Furthermore, by using the PMT housing as a light guide to reduce the photocathode area, the dynode assembly can be made smaller and the light more evenly distributed.

The large 130 mm PMT exhibits substantial anode response nonuniformity induced by variable photon and photoelectron transit. A reflective photocathode reduces nonuniformity from QE-enhancing back-reflection, as the metallic photocathode substrate has uniform reflective properties. Additional nonuniformity caused by electron transport to the dynodes may be mitigated since the dynodes stretch the full photocathode length, leaving the sub-50 mm photocathode cross-sectional width as the primary contribution to incident-trajectory variation rather than the 130 mm diameter of the 9490B.

Data provided by these CsI(Tl) spectrometer measurements will provide validation models for accurate Monte Carlo simulation of the gamma/optical-photon responses of both commercial and novel scintillation spectrometers. Ongoing studies of identically-oriented $25.4 \times 127 \times 25.4 \text{ mm}^3$ NaI(Tl) crystals will provide more validation data for different reflectors, surface morphologies, and optical couplings. Similarly, spatial measurements using a $50.8 \times 152.4 \times 50.8 \text{ mm}^3$ NaI(Tl) crystal will validate models of large-volume crystals paired to the commercial 9490B PMT and enable simulation of novel photodetector concepts.

Chapter 3

PMT Simulation Development and Novel PMT Design

An array of PMT designs have been developed and characterized through the technology's 90-year existence. The current work seeks to integrate cathode and dynode design elements which have historically provided good timing and response uniformity. An iterative design process was performed to model and adapt well known discrete dynode designs to the rectangular PMT form factor. To simulate the novel PMT design, phenomenological models of photo- and secondary electron emission and transport were developed and validated using data from commonly-used, well-studied sensitive materials and devices.

3.1 Sensitive Material Selection

The energy resolution of a scintillation spectrometer is dictated by the signal variance as a consequence of the chosen emissive materials, operating potential, and detector geometry. For example, variance in the number of scintillation photons produced per interaction is intrinsic to the material, whereas variance in the photon transfer efficiency is the result of optical materials (crystal, reflectors, windows, etc.) as well as the housing and optical interface geometries. It is imperative to maximize the number of photoelectrons produced

and collected so that the effect any single photoelectron has on the variance of the total signal is reduced. Hence, a large rectangular optical interface was chosen to maximize the optical coupling area. Similarly, the sources of variance in the amplification process must be limited. To improve both the transfer and multiplier resolutions, one must use high-performance electron emitters for the photocathode and dynodes. Commercial manufacturers often turn to alkali-antimonides such as Cs_3Sb and K_2CsSb ¹⁶, which offer great performance as both photoemitters and secondary emitters, as shown in Figs. 1.17 and 1.20. Using the same or similar materials for both also simplifies fabrication. The sections which follow offer a comprehensive look at how the emissive properties of these two materials were modeled to enable studies of PMT performance.

3.2 Simulation of Electron Transport and Multiplication

Charged particle transport and multiplication in electric and magnetic fields have successfully been simulated using multiphysics Finite Element Method (FEM) codes such as COMSOL, CST Particle Suite, and SIMION^{75–81}. In FEM, a geometry is discretized into point nodes which represent pertinent physical properties or quantities at a specific location. Adjacent nodes are linked by elements representing an approximation of the partial differential equation used to describe the appropriate physics. The combination of nodes and elements distributed throughout a defined geometry is known collectively as the mesh. For these studies, a second-order tetrahedral mesh was used as the basis of the simulation. Quadratic mesh elements are selected by default and were chosen to balance simulation speed and accuracy for the many curved surfaces. One such example for the simulated ET Enterprises 9214B PMT may be seen in Fig. 3.1.

Within COMSOL Multiphysics®, the chosen simulation software, vertices, edges, surfaces, and domains are defined by the geometry components. Volumes, or domains, are assigned material properties, while surfaces provide the boundary conditions necessary to

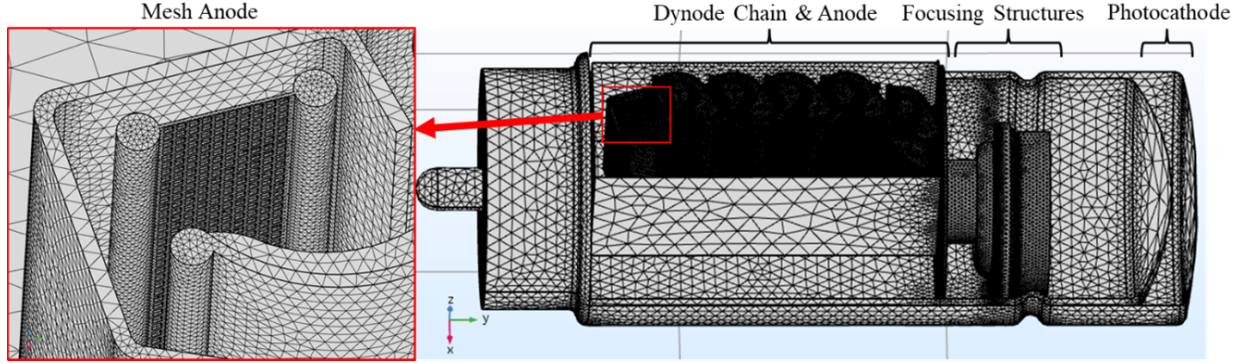


Figure 3.1: *The tetrahedral mesh used to simulate the ET Enterprises 9214B PMT which served as the model validation.*

reach a solution. For charged particle simulations described, a static electric field — unchanged by the charges being transported — was first simulated, followed by the electron transport described in following sections.

3.2.1 Electrostatic Simulation

Electrostatic simulation using the COMSOL AC/DC module was relatively straightforward. A mesh of the vacuum domain defined the volume in which the electric field distribution was calculated. Electric potential applied to the surfaces comprising each electrode and the photocathode served as the boundary condition. Glass substrates with no conductor maintained the default “Zero Charge” definition. The electric field is solved by approximating Gauss’s law

$$\nabla \cdot \mathbf{E} = \frac{\rho}{\varepsilon_0}, \quad (3.1)$$

where ρ is the charge density, and ε_0 is the vacuum permittivity. The electrostatics solution was then fed into the charged-particle solution using the Electric Force node under the Charged Particle Tracing physics header.

3.2.2 Electron Transport

The same mesh used to calculate electrostatic solutions was used for time-dependent Charged Particle Tracing in COMSOL. The charged particle was defined as an electron using the

default mass and charge settings and was tracked using the standard equations of motion. As previously stated, the electric force applied to each electron was calculated via the Lorentz force equation

$$\mathbf{F} = q_e \mathbf{E}. \quad (3.2)$$

The static electric field $\mathbf{E}$ is imported from the separate electrostatic study. No consideration was given to electron interactions in the vacuum, nor was space-charge buildup considered over the range of simulated gain. This is consistent with other work^{82;83}; however, space charge buildup at the anode may be significant in some high-gain or high photon-flux applications. Its inclusion is left for future work, if deemed necessary.

Primary particles (photoelectrons) were introduced at the photocathode surface. Electrode boundaries were set to “Freeze” or “Disappear” particles as needed. Secondary particles were produced at dynode surfaces. The release properties of each are described in great detail throughout the following sections.

3.2.3 Photoelectron Emission

During electron transport simulations, photoelectron release energies were pseudorandomly sampled from a distribution derived from the convolution of three other material-specific distributions. This process is adaptable, and data from alternative scintillators or photocathodes may be substituted if needed. First, a typical NaI(Tl) emission distribution was convolved with the energy-dependent QE of a typical K₂CsSb photocathode to produce a wavelength-dependent photoelectron emission probability distribution as seen in Fig. 3.2.

Data from Nathan and Mee²⁵ pertaining to wavelength-specific photoelectron emission energies were fit using the skew- t distribution, as plotted in Fig. 3.3. The skew- t distribution was chosen due to its versatility, utilizing 4 variables which provide control over the first four central moments: mean, variance, skew, and kurtosis⁸⁴. A pre-built function in R statistics programming language was used to produce individual skew- t distributions for monochromatic light at a few wavelengths.

The original, continuous convolution spectrum derived from emission and absorption

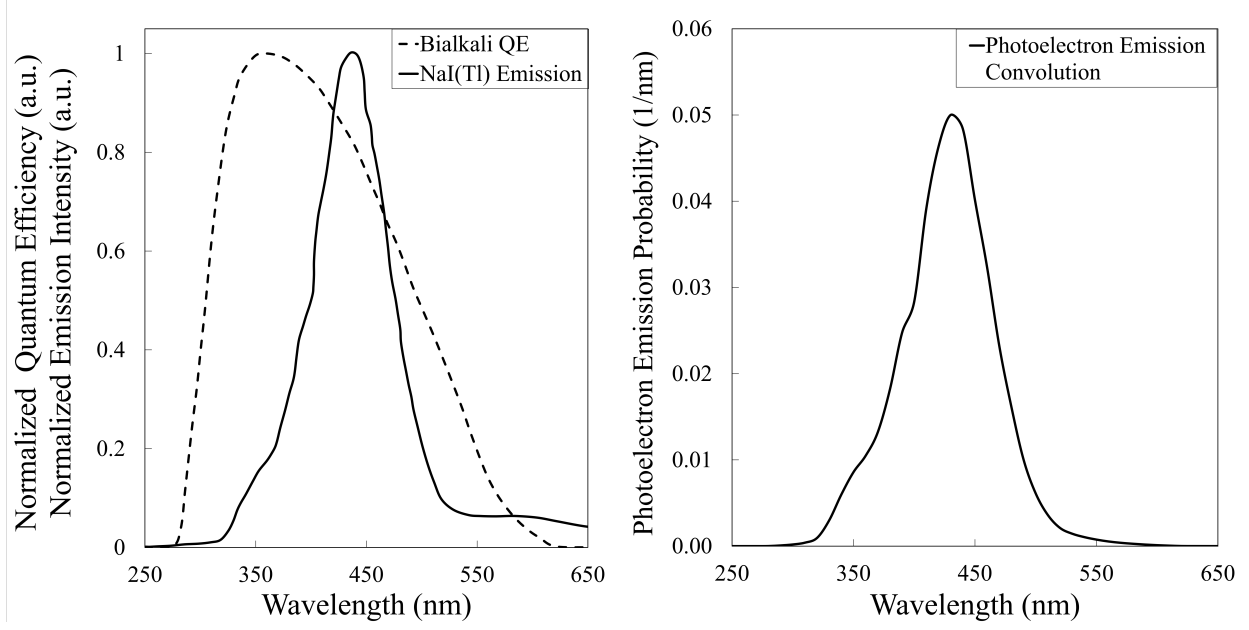


Figure 3.2: *Left) Normalized QE of K_2CsSb photocathodes and normalized $NaI(Tl)$ scintillator light emission intensity^{23;24}. Right) The resultant wavelength-dependent photoelectron emission probability distribution for a K_2CsSb photocathode paired to a $NaI(Tl)$ scintillator.*

data was discretized in 10 nm wavelength intervals from 290 nm to 650 nm producing a probability mass distribution of the absorption spectrum at the photocathode. The four parameters for each skew- t distribution produced by fitting Nathan and Mee’s data were linearly interpolated to produce intermediate curves for each of the discrete wavelength intervals. No data was available for lower-energy photons, thus data was extrapolated out to the low-energy bound of the photocathode absorption spectrum (1.9 eV, in this case). Given the near-zero probability of photon absorption at energies < 2.25 eV (550 nm) and the low photoelectron energies produced, systematic error arising from extrapolation is assumed to be negligible. Similarly, imperfect fits to photoelectron energy distributions displaying additional spectral features for photon energies $> 2E_T$ (325 nm) — caused by the production of a second photoelectron — were deemed immaterial, since very few photons emitted from $NaI(Tl)$ and $LaBr_3(Ce)$ scintillators are of sufficient energy.

By convoluting the discretized absorption distribution and photoelectron energy distributions, a full photoelectron emission spectrum was produced, and can be seen in Fig. 3.4. A binary output file was produced containing approximately 1,500,000 pseudorandom

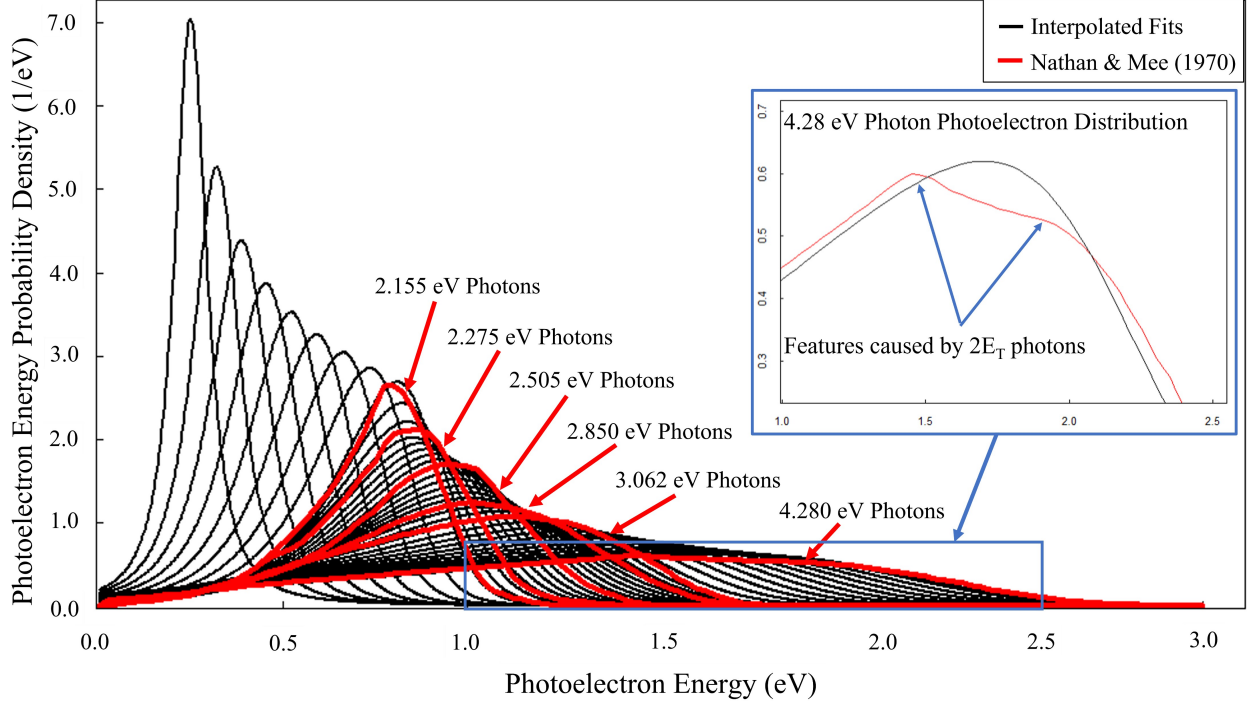


Figure 3.3: Photoelectron energy distributions produced by monochromatic light²⁵ (red) were fit using the skew-t distribution. After the initial distributions were fit, the four variables were linearly interpolated to provide a 10 nm discretization. Additional features in the 4.28 eV-photon photoelectron energy distribution stem from photons with energies in excess of $2E_T$ producing two photoelectrons per absorption.

photoelectron energy samples. An external C subroutine is called by COMSOL to provide pseudorandom samples of the photoelectron energies contained within the file.

The result is a distribution with a mean value of 0.96 eV and a FWHM of 0.74 eV, which is similar-to but broader-than distributions previously used by the author⁷⁵ and others who have used a fixed emission energy of 1 eV¹⁸. For the current work, the minor dissimilarity between these photoelectron emission spectra primarily affects photons emitted near the cathode perimeter which are subjected to low extraction fields. However, this technique may also be used in future work for scintillators which emit higher-energy photons and, subsequently, higher-energy photoelectrons that may overcome low extraction fields. For the current studies, photoelectrons were released following a Lambertian cosine directional distribution, as is common among other texts¹⁸

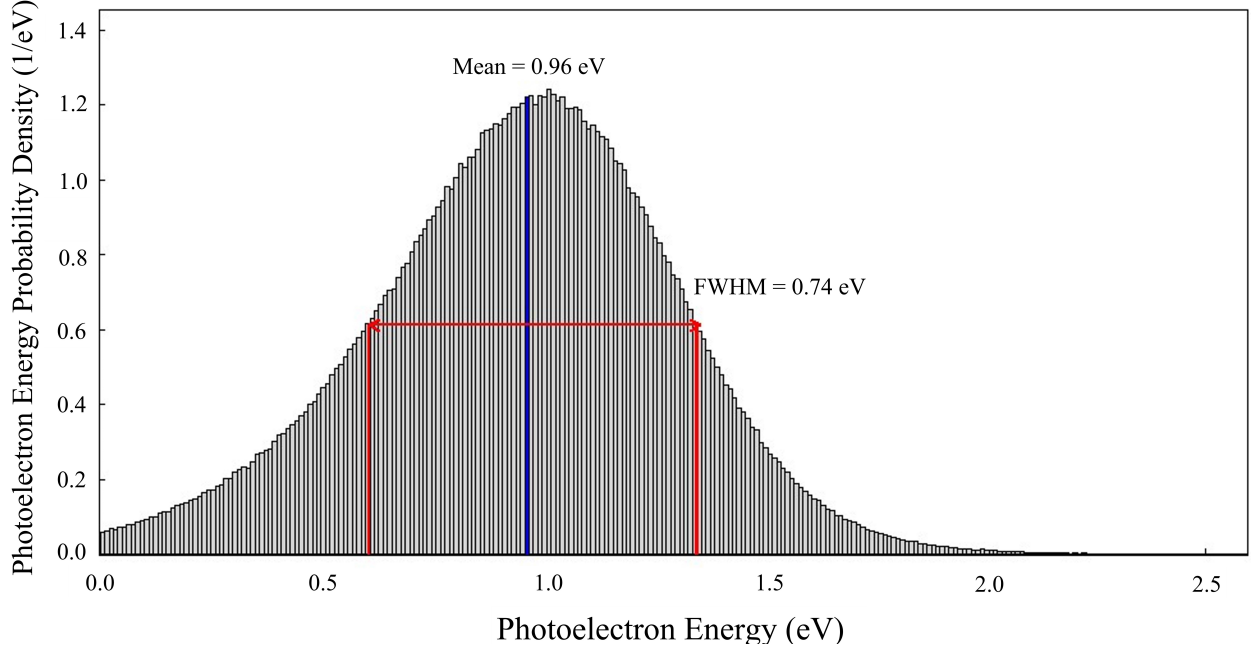


Figure 3.4: *The final sampled photoelectron energy distribution which was saved to a binary file and later pseudorandomly sampled by an external subroutine called by COMSOL.*

3.2.4 Secondary Emissive Yield

Both modern and historical texts have used different terms to describe the average ratio of emitted electrons to the number of incident primary electrons. This ratio is known as both the total electron yield (TEY) and, more casually, secondary emissive (or electron) yield to describe the same parameter⁶⁵. Many works have tended towards SEY, which is likely due to the relatively high yield of true secondary electrons compared to scattered electrons in many materials. TEY will be referred to as the more commonly used term, SEY, following the notation of Dekker⁸⁵, and the ratio of *true* secondary electrons to the number of incident electrons will be distinguished as the “true secondary electron yield”. The total SEY is given by

$$\delta_{tot} = (1 - \eta)\delta_{ts} + \eta\delta_{bs}, \quad (3.3)$$

which is a summation of the fraction of primary electrons *not* scattered $(1 - \eta)$ times the true secondary electron yield from those primaries, δ_{ts} , and the backscatter electron yield, δ_{bs} times backscatter coefficient, η . The backscatter coefficient, η , is simply the ratio of reflected

electrons (RE) to incident primary electrons (PE).

Secondary Electron Yield for Normally-Incident Primary Electrons SEY is well defined for various materials in multiple texts^{19;85–87}. Energy-dependent SEYs for Cs_3Sb and K_2CsSb were digitized from Sommer’s data¹⁹, which may be seen in Fig. 3.5. A simple cubic fit was applied to the data for each. Dependent upon the simulated material, COMSOL first calculates the deterministic, energy-dependent SEY based on the provided fit. The SEY is then modified and sampled following the processes outlined in the following sections.

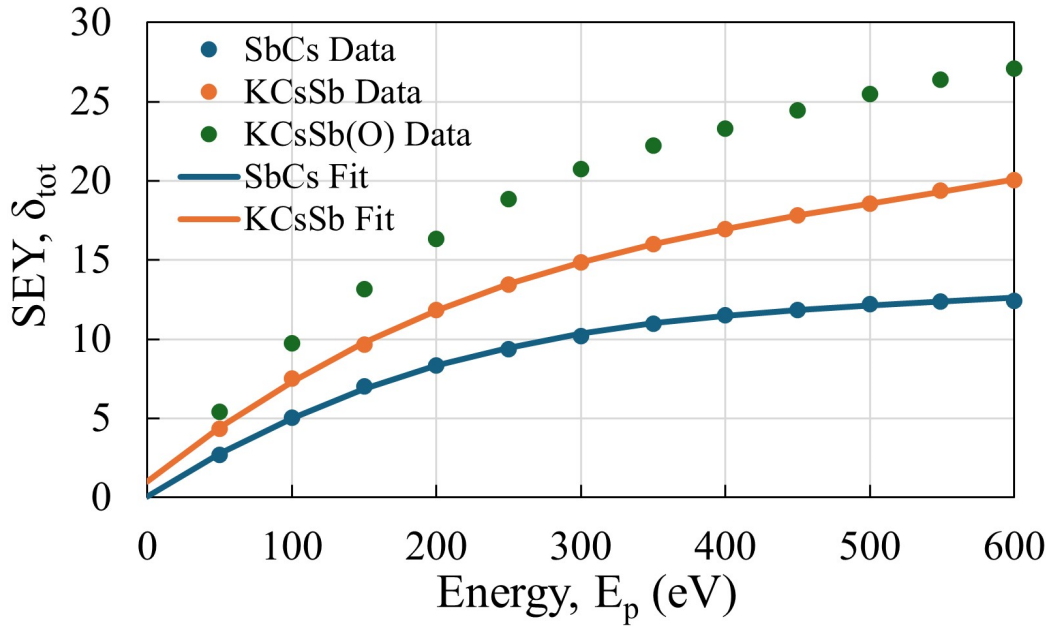


Figure 3.5: Measured energy-dependent total SEYs at normal incidence for a few alkali-antimonide secondary emitters are provided¹⁹. Cubic fits (solid lines) to data for Cs_3Sb and K_2CsSb were used for the simulations described herein. Note that a superficial oxidation layer on the surface of the bialkali material enables more efficient secondary emission by increasing the escape probability due to band-bending at the surface.

Dependence on Angle of Incidence It has been shown that increasing the angle of incidence with reference to the surface-normal increases the SEY^{26;64;85;88;89}. The backscatter coefficient is similarly affected²¹. This is intuitively understood, as secondary electrons produced near the emitter’s surface have a lower probability of absorption as they travel to the surface. The primary electron angle of incidence, therefore, is an important parameter to

incorporate. Consider a secondary electron excited by a primary electron that has traveled a depth $x = l \cos \theta$ into the material. The secondary electron has a probability of being reabsorbed which increases with distance r from the interaction location. For increasing angles of incidence, θ , the depth of interaction is reduced, meaning the distance that must be travelled to the surface (and the probability of reabsorption) for a secondary electron is reduced. Hence, the probability of escape is increased. This is illustrated in Fig. 3.6. Due to

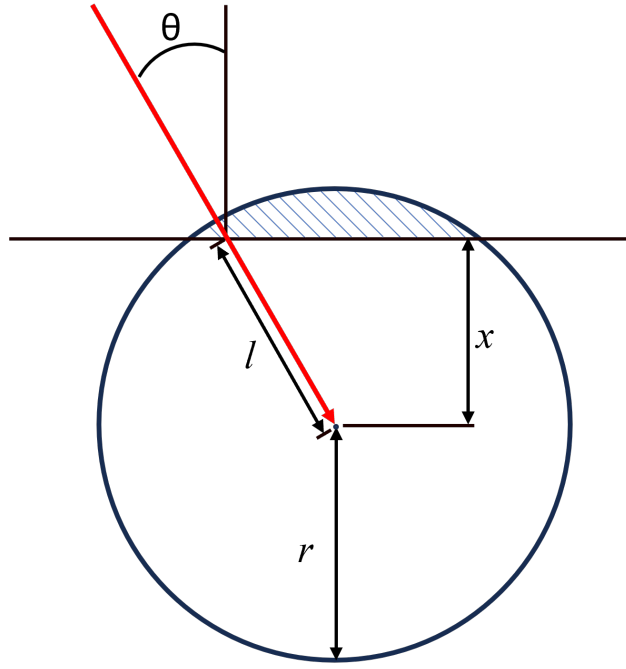


Figure 3.6: *The angle of incidence affects the number of secondary electrons emitted, since there is an increased probability of producing secondary electrons near the surface for higher angles of incidence. A secondary electron (red) travels some distance l into the material and interacts at a depth x from the surface. The probability of reabsorption increases with distance r from the interaction, meaning electrons produced closer to the surface have a higher probability of escape.*

the complex processes of electron energy-deposition and emission, the yield cannot simply be multiplied by a factor of $1/\cos \theta$. Indeed, many scientists have suggested modifications to the normal-incidence SEY to model the effects of incidence angle considering both the material topography and crystallinity^{21;65;87;90}. Bruining's formula,

$$\frac{\delta(E_p, \theta)}{\delta(E_p, 0)} = e^{\gamma(1-\cos \theta)}, \quad (3.4)$$

was used to modify the SEY as well as the backscatter coefficient in the current work. The material parameter γ is a semi-empirical term related to electron absorption coefficient and diffusion range. This value was applied separately to both SEY and the backscatter coefficient.

No γ values were found for alkali-antimonide based secondary emitters, but it is understood that values typically lie between 0.2-0.7²¹. Several individual fits derived from $\delta_{tot}(E_p, \theta)$ data for Mo were used, as the atomic number of Mo ($Z = 42$) is reasonably close to that of K₂CsSb ($Z_{eff} = 44$)^{26;87;88}. Following the extensive data provided by Shih²⁶ (data points in Fig. 3.8), one finds that γ is energy-dependent and may be fit using

$$\gamma(E_p) = a(1 - e^{bE_p}), \quad (3.5)$$

where $a = 0.71$ and $b = -0.0039$ are fit parameters. The $\gamma(E_p)$ fit function is plotted in in Fig. 3.7 alongside the results of several individual $\gamma(E_p)$ fits.

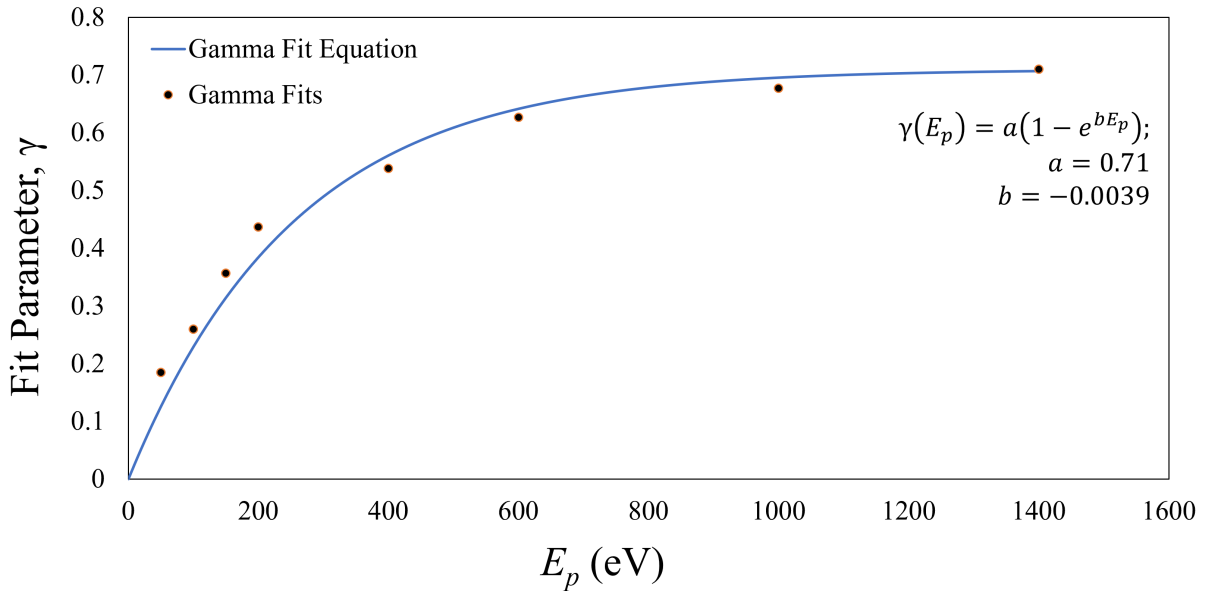


Figure 3.7: Using Mo SEY data from Shih²⁶, a function describing $\gamma(E_p)$ was derived.

$\delta(E_p, \theta)/\delta(E_p, 0)$ as estimated by the $\gamma(E_p)$ fit-function is shown for several primary-electron energies in Fig. 3.8. The $\gamma(E_p)$ fit-function is conservative ($< 10\%$ different) at low energies for high angles of incidence compared to the individual fits used to derive it.

Discrepancies between the individual fits for $\gamma(E_p)$ and the $\gamma(E_p)$ fit equation for $E_p < 600$ eV may justify a modified equation to accommodate a y-intercept > 0 , although this was not included in the current work. Alternatively, an interpolation table composed of individual $\gamma(E_p)$ fits may be imported to COMSOL, if desired.

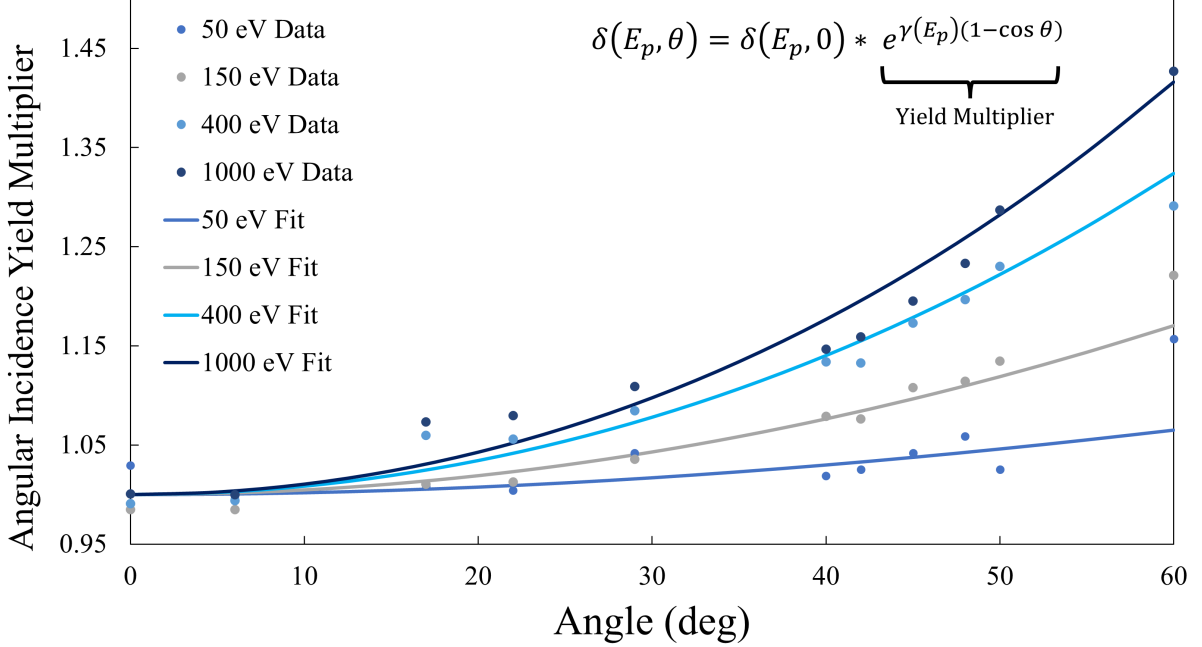


Figure 3.8: Energy and angle-dependent fits to the “yield multiplier”, $\delta_{tot}(E_p, \theta)/\delta_{tot}(E_p, 0)$, using the γ fit-function and parameters shown in Fig. 3.7. At higher angles of incidence ($\theta > 50^\circ$), the differences between the individual $\gamma(E_p)$ fits and the $\gamma(E_p)$ fit-function at low-energies are apparent.

One may recall that three types of secondary electrons (true, rediffused, and elastically-scattered) are considered and may be differentiated by their energy spectra. To sample yield, secondary electron emission is initially considered binary, with the two options being non-backscattered (true secondaries) and backscattered (all others). The backscatter coefficient is dependent upon the effective atomic number of the emissive material, which can be found using

$$Z_{eff} = \frac{mZ_A^2 + nZ_B^2 + \dots}{mZ_A + nZ_B + \dots} \quad (3.6)$$

for any compound A_mB_n ¹⁸. Rectangular PMT simulations use K_2CsSb as the secondary emissive material ($Z_{eff} = 44$), while data are provided for electrons impinging Cs_3Sb ($Z_{eff} =$

54) and NaKCsSb ($Z_{eff} = 34$) at normal incidence²⁷. Figure 3.9 shows the experimentally measured backscatter coefficients and fits to pertinent materials. The fit equation

$$\eta(E_p) = r_1 (1 - e^{r_2 E_p + r_3}), \quad (3.7)$$

uses 3 fit parameters, r_1, r_2 , and r_3 , which were found through non-linear least-squares regression of the provided data.

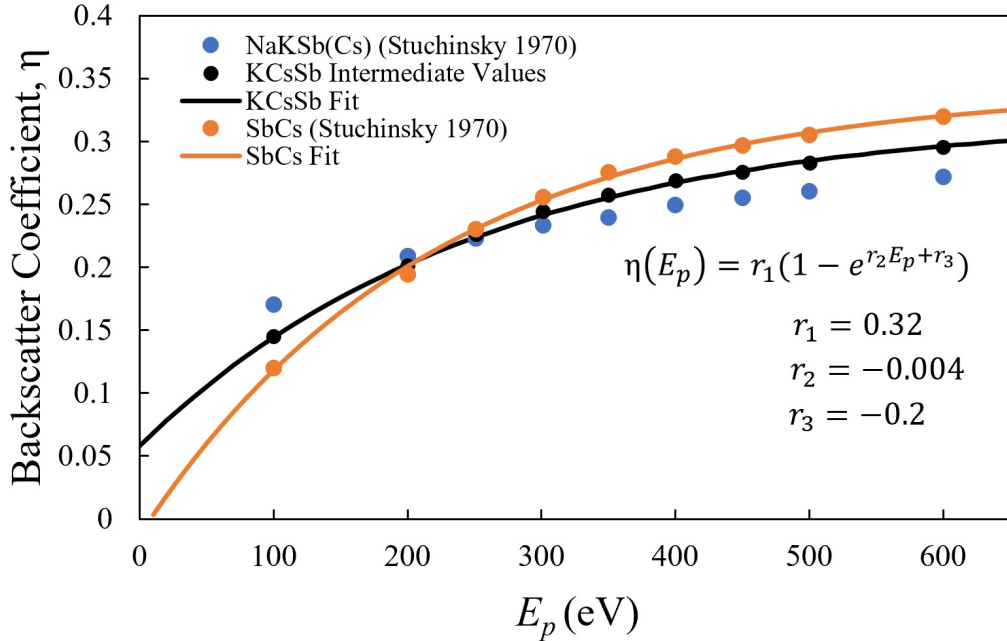


Figure 3.9: The normal-incidence backscatter coefficient fit for two alkali-antimonide secondary emitters (NaKSb(Cs) and SbCs)²⁷, as well as the fit to the interpolated backscatter coefficients for K_2CsSb . The fit provided is for K_2CsSb . Fit parameters for Cs_3Sb are: $r_1 = 0.343$, $r_2 = -0.0046$, $r_3 = 0.0366$.

To calculate the energy *and* angle-dependent backscatter coefficient, $\eta(E_p, \theta)$, a fixed $\gamma = 0.33$ ²¹ was used. This γ value has no energy dependence and was simply taken from emission models of 304 stainless steel and Cu. Secondary emission studies are primarily focused on electron microscopy applications, thus, to the author's knowledge, no data is available for alkali-antimonides. Since the overall backscatter yield is relatively low, especially for high-yield semiconductors, this rough approximation was deemed sufficient. Below, in Fig. 3.10,

the average simulated energy and angle-dependent SEY and backscatter coefficients are plotted for Cs_3Sb and K_2CsSb .

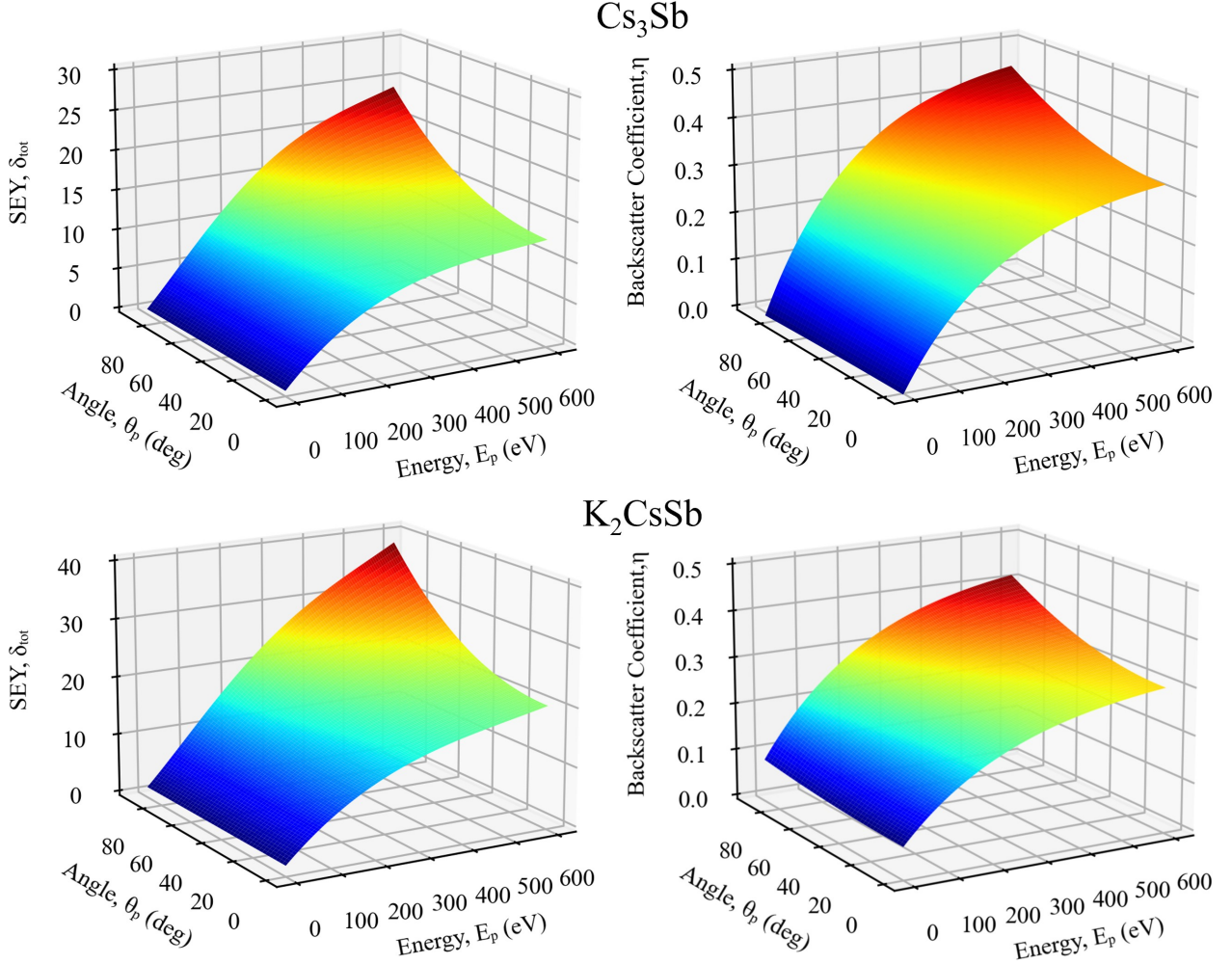


Figure 3.10: $\delta(E_p, \theta)$ and $\eta(E_p, \theta)$ are provided for the alkali-antimonide secondary emissive materials used for the simulated dynodes. The color scale is normalized by the maximum value for each individual plot.

For elastic and inelastic scattered electrons – together comprising backscatter – the yield was simply considered to be one. Therefore, the true secondary electron yield may be calculated as

$$\delta_{ts} = \frac{\delta_{tot} - \eta \delta_{bs}^{\rightarrow 1}}{1 - \eta}. \quad (3.8)$$

Using the calculated true secondary yield as the mean, an external C function was used to pseudorandomly sample the Poisson distribution. Although the Furman model uses a

Binomial distribution for additional control over the variance, the Poisson distribution is suggested as a viable alternative to determine a discrete number of secondary electrons²¹. Since no data has been collected on the variance of the yield for K_2CsSb or similar materials, it was decided that the Poisson distribution may be considered a reasonable assumption.

Although the assumption that backscattered electrons produce a single secondary electron is true for elastically scattered electrons which lose minimal energy in their interaction, inelastically scattered electrons impart much more energy to electrons within the material. This means that rediffused electrons have a higher probability of also producing true secondaries⁶⁵. True secondary electron emission was not considered during backscatter events in the current model due to difficulty in returning an array of associated values (e.g. yield and energy) to COMSOL from an external code. Instead, additional true secondary electrons are emitted by the standard true-secondary condition to recover the difference. The primary deficiencies of this process are the loss of mid-level single-electron responses and the positional-displacement of some true secondary electrons. In other words, a simulated true-secondary emission will produce a higher yield than in actuality, and conversely, simulated inelastic-scatter events will have a yield of just one — lower than that observed by measurement. While the overall gain will remain the same, one should expect fewer medium-sized responses and an excess number of low-level responses in the SER distribution. This assumption follows a similar mutual-exclusion clause found in Furman’s work²¹; although, future work should seek to remedy the issue.

3.2.5 Secondary Electron Energies

The secondary electron energy spectrum follows the generalized shape previously discussed in 1.5.4. In this work, a simplified form of the Furman model of secondary electron emission was implemented within COMSOL. Separate models were implemented for each of the three primary spectral features.

True Secondaries An approximation of the secondary electron energy distribution given by Bateman⁸² was used for both K₂CsSb and Cs₃Sb:

$$\frac{dN}{dE_s} = \begin{cases} \frac{a}{E_s} & 0\text{eV} < E_p < 8.5\text{eV} \\ a \left(e^{-\frac{E_s}{t}} - e^{-\frac{E_s}{1}} \right) & 8.5\text{eV} \leq E_p < 40\text{eV} \\ a \left(e^{-\frac{E_s}{5}} - e^{-\frac{E_s}{1}} \right) & 40\text{eV} \leq E_p < \infty. \end{cases} \quad (3.9)$$

Values were rejection sampled from these distributions by calling an external C subroutine. For primary electron energies above 40 eV, the standard fit was used. For E_p between 8.5 eV and 40 eV, the distribution was scaled using Eq. 3.10

$$l = \frac{6(E_p - 42.5)}{E_p}, \quad (3.10)$$

where l is a scaling factor used to limit the probability of true secondary electron energies sampled above the primary electron energy. Below 8.5 eV, electron energies are uniformly sampled from 0 eV to the primary energy threshold. There is no physical basis for scaling true secondary electron energy distribution this way. It was simply used to reduce the probability of sampling a secondary electron energy greater than that of the primary electron. Future work may simply limit the bounds of the distribution provided by Bateman, or should COMSOL allow, develop a coherent emission model such that $\sum_{i=1}^n E_{s,i} \leq E_p$.

Rediffused Primaries The next distribution, rediffused electrons, represents those primary electrons which impart some of their energy to other electrons in the bulk material, but ultimately escape with some fraction of their original energy remaining. Energies comprising this distribution are uniformly sampled over the range $[0, E_p]$, with a mean value of $0.5E_p$. Typically, this region of a measured spectrum is slightly concave, but other works have found this to be an unnecessary addition to the model given its relatively small contribution¹⁸.

Elastically Scattered Primaries The third and final region is composed of backscattered primary electrons. Backscattered primary electrons maintain most of their incident energy, and are modeled as a Gaussian distribution with a mean energy, $\mu = E_p - 2\sigma$, and a standard deviation, $\sigma = 0.005E_p$, though given the small energy spread associated with reflected electrons, an emission energy of E_p would have been sufficient.

For the purposes of modeling the full energy distribution, backscattered electrons over the range $[0, E_p]$ were further separated into two categories: rediffused electrons and elastically-scattered electrons — each having a distinct probability of occurring. Following a coarse approximation provided by Zahid (p. 56)⁸³, the probability of an elastic scatter was assumed to be a fixed 13.8% of all backscatters, even though this ratio is understood to vary dependent upon energy and angle^{21;85}. For example, Fig. 3.11 illustrates measured true secondary, rediffused, and elastic scatter yields for stainless steel at normal incidence.

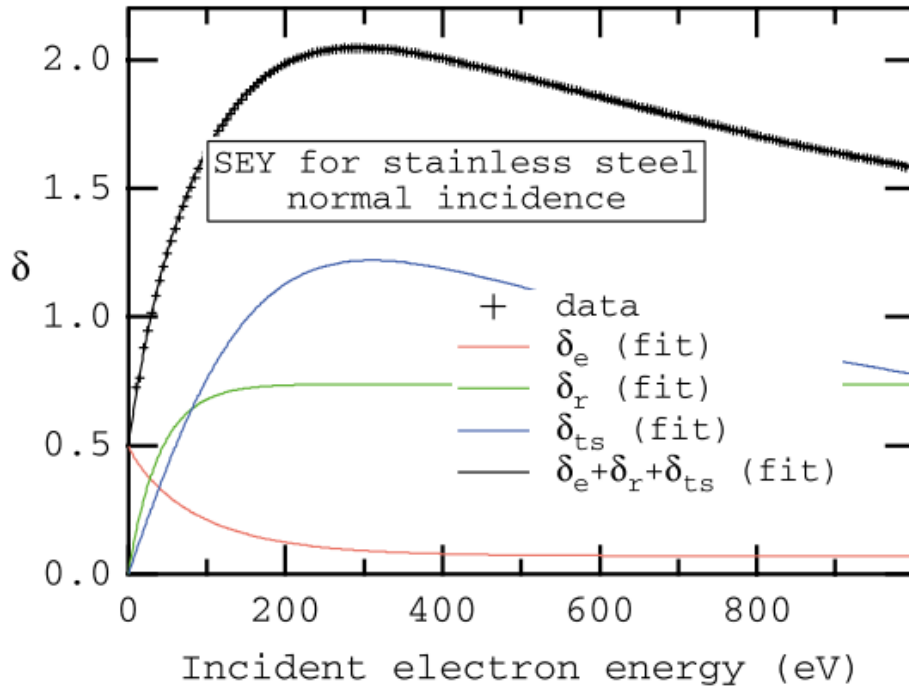


Figure 3.11: *Relative yields of the three types of secondary emission considered for stainless steel from Furman²¹. Data provided by R. Kirby (unpublished). At low energy, elastic scatter is the primary emission mode. True secondary emission dominates at higher energy due to the greater number of electrons produced per event.*

Full Secondary Electron Energy Distribution Combining the yields and energy distributions of each described secondary electron emission modality results in the full secondary electron energy distribution. This distribution is unique for any incident primary electron energy and angle. An example distribution, sampled using COMSOL, is provided in Fig. 3.12 for normally-incident electrons at 300 eV. Angle of incidence was not considered when sampling individual secondary electron energy distributions.

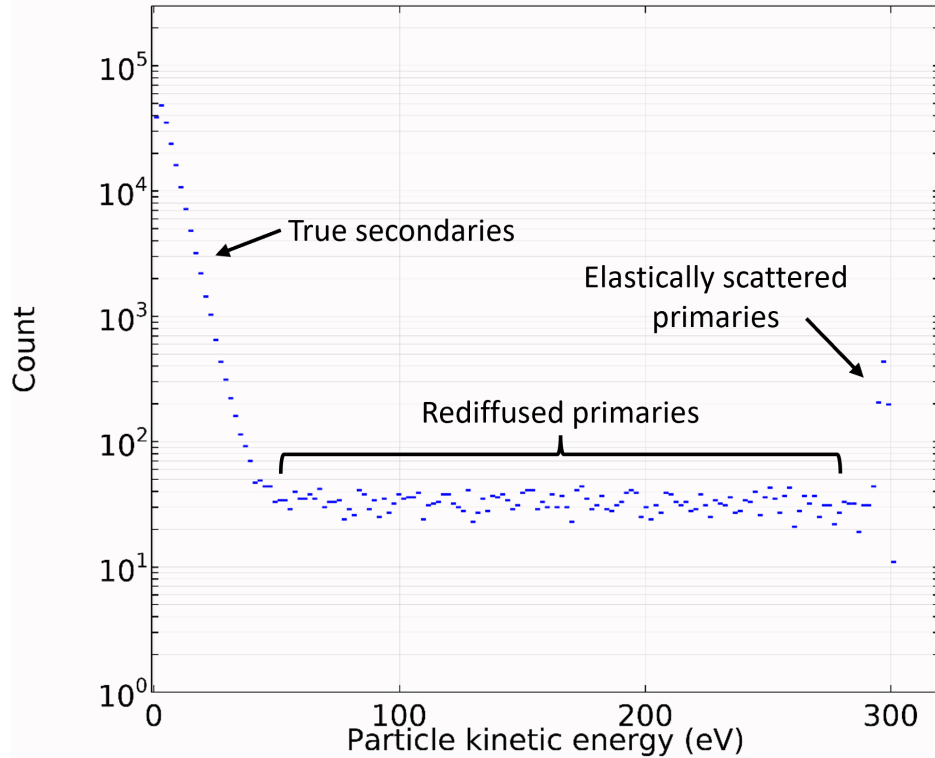


Figure 3.12: *The secondary electron energy spectrum for SbCs as sampled by COMSOL for 300 eV, normally-incident primary electrons. The ratio of the integrated backscatter response to that of the true secondary electrons is modified by the secondary yield and backscatter coefficient for emission from KCsSb.*

The angular distribution of true secondary electrons and rediffused electrons are generally accepted to have a Lambertian (cosine) distribution^{18;82;85}, and backscattered electrons followed the same assumption for simplicity.

3.3 Emission Model Validation

3.3.1 Geometric Modeling

An extensively characterized commercial photomultiplier was chosen to validate the proposed emission models. An ET Enterprises 9214B PMT was obtained and reconstructed for simulation. The 9214B has a 51 mm diameter bialkali (K_2CsSb) transmission photocathode. It has 12 Cs_3Sb linear focus dynodes and a wire-mesh anode. Projectional radiograph images were taken of the ET Enterprises 9214B PMT using a Philips SRO 33-100 system operated at 60 kVp, which provided high resolution for small features but did not provide 3-dimensional information. Computed Tomography (CT) scans were taken with a GE BrighSpeed 16-slice machine, which were effective in reconstructing the full geometry but could not resolve small details — such as the wire-mesh anode — with the accuracy provided by the projectional radiograph. Utilizing both imaging techniques, a relatively accurate physical model was created using Solidworks. Figure 3.13 shows the modeled geometry (orange) overlaid atop a cross-sectional image of the 3D model produced via CT. Combined, the imaging methods

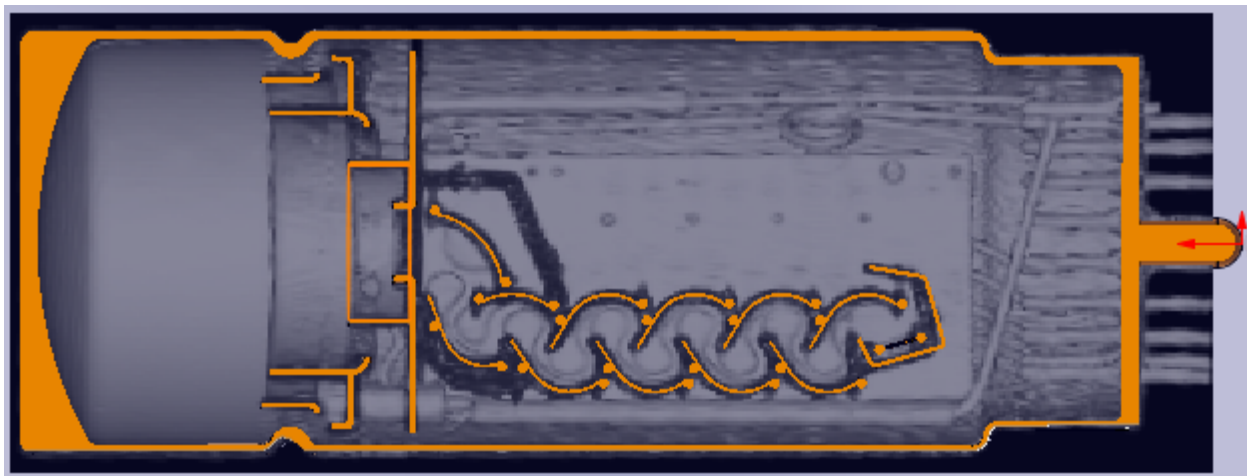


Figure 3.13: *A cross-section of the 9214B PMT geometry recreated in Solidworks is shown in orange, while a cross-section of the 3D model produced via CT may be seen in the background. Some features outside of the electron-transport region were left unmodeled.*

captured nearly all supporting structures and wire connections within the PMT; however, only surfaces pertinent to electron transport were recreated in the CAD geometry. This

includes only the electrode surfaces and the outer envelope, thereby reducing the number of mesh elements and computational time required for simulation. The simplified model was imported directly to COMSOL for physics modeling. Further, minor modification of the dynode chain was informed by simulation and performed iteratively in COMSOL. This was required to enhance the simulated interstage electron-optics and approximate experimental results.

3.3.2 Electrostatic Simulation

To begin, an electrostatic simulation of the full model was performed. Table 3.1 is the voltage distribution used throughout the 9214B simulation process. One such solution may be seen

Table 3.1: *The voltage distribution used for 9214B electron transport studies²⁴. K , D_n , and A represent the cathode, n^{th} dynode, and anode, respectively. A fixed 300 V is applied from K - D_1 , and R indicates an equivalent resistance value used to distribute the K - A operating potential amongst the dynodes.*

K	D_1	D_2		D_{11}	D_{12}	A
300	R		R	2R	R	

in Fig. 3.14. The electrostatic solution was then imported to the electron-transport study, as described previously.

3.3.3 Charged Particle Transport Methods

Different techniques were used to more efficiently simulate electron transport and multiplication within the validation model dependent upon the measured characteristic. To quantify SER, 2600 studies were performed. For each, an individual photoelectron was released. The full photoelectron and secondary electron release processes were included, and the resultant cascade was tracked until the sixth dynode. From there, the results were numerically cascaded to the anode following a Poisson SEY condition using a Python script.

For timing studies, backscatter was “turned off”, and SEY was reduced by one-third to reduce the computation time. 1300 photoelectrons were released simultaneously from the photocathode and tracked for 12 ns, such that none reached the first dynode. At 12 ns,

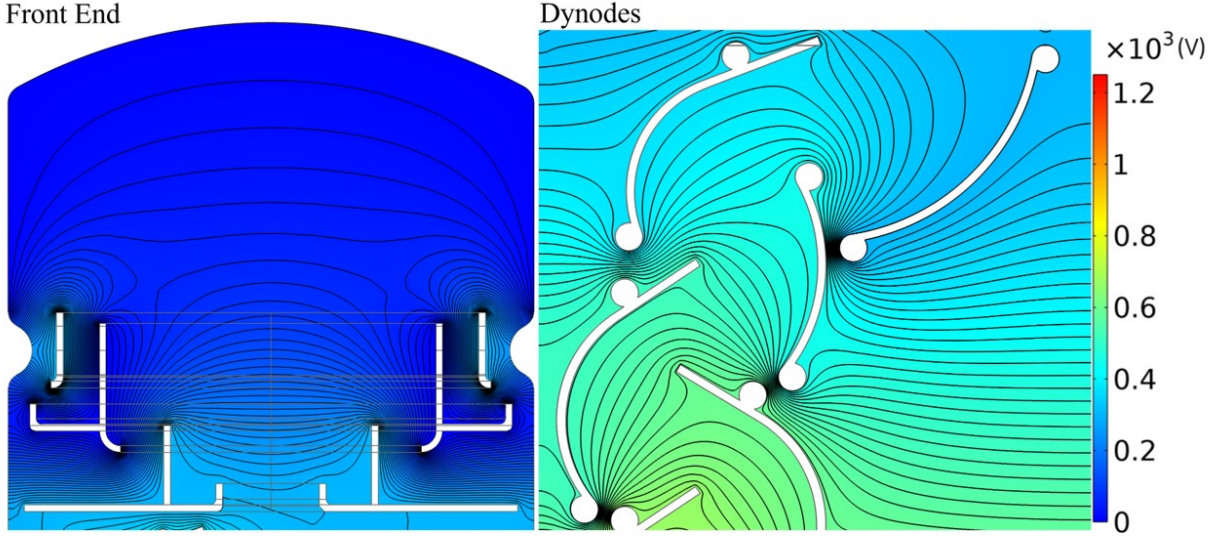


Figure 3.14: *The 1250 V operating potential solution for the 9214B PMT with separate emphasis on the front end (left) and the first few dynodes (right). The colormap represents electric potential, while black curves reveal 10 V/div equipotential lines.*

the positions and velocities were recorded in separate files. A parametric study was used to continue the simulation for each individual electron from 12 ns. Electrons cascaded through the dynodes and were collected at the anode and the cumulative charge was recorded in 0.25 ns intervals. Total simulated time for each solution, including the initial 12 ns, was 65 ns. The electron collection time-distribution was differentiated to calculate the rate of charge collection which served as an analogue to current. It should be noted that COMSOL's built-in current probe is tied to a current release definition, which cannot be used for single-electron releases. Implementing a custom study to solve the Shockley-Ramo theorem for induced charge, while possible, would require an additional solution for the weighting-field and computation of the induced charge at each time-step lengthening the simulation time. Figure 3.15 represents a typical (partial) charged-particle tracking solution for the 9214B in COMSOL.

3.3.4 Results

Single Electron Response The single electron response was simulated to the sixth dynode to minimize computation time, while ensuring accurate simulation of the PMT front

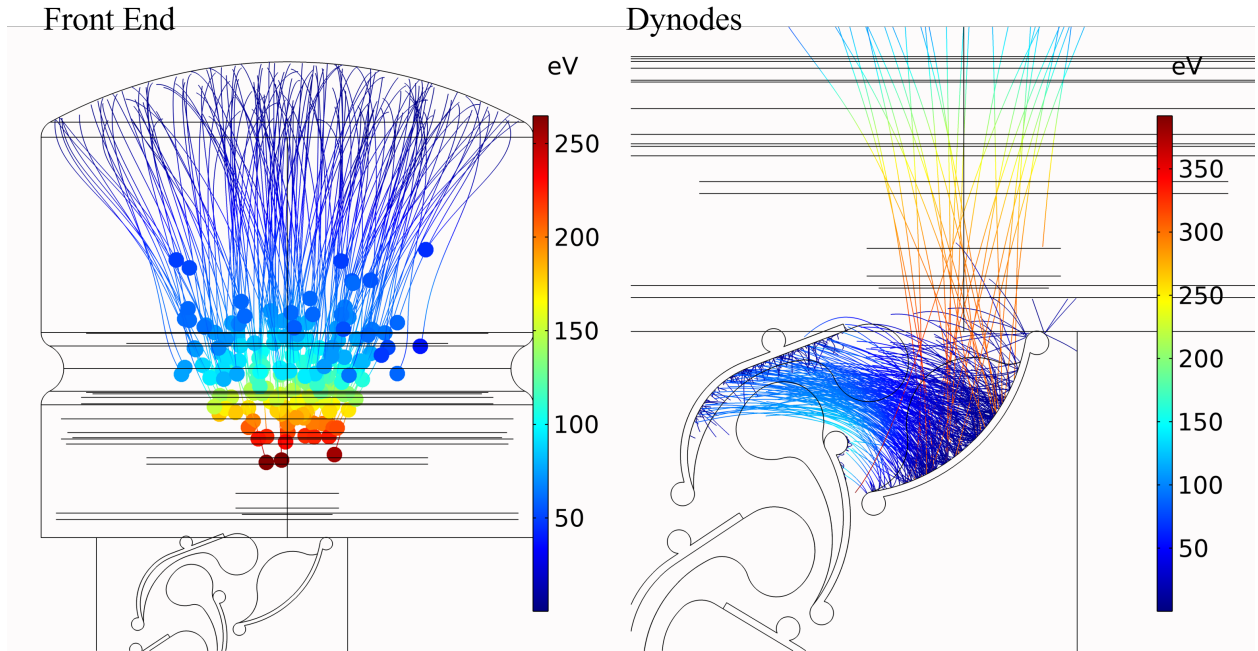


Figure 3.15: A partial charged-particle tracking solution showing the front end (left) and first few dynodes (right) illustrates how photoelectrons are focused and accelerated to the first dynode. Interdynode transport reveals some losses due to unfavorable interaction locations or initial trajectories.

end and “transition” dynodes (1-3). These first few dynodes have the largest statistical effect on SER due to variance in the SEY (as described in Section 1.5.5) and due to reduced interstage CE as the dynodes transition to the optimized linear-focus geometry. Beyond the sixth stage, a Python script was employed to calculate SER at the anode. The multiplication process was modeled following a simple Poisson distribution of SEY, ignoring backscatter, but considering realistic interstage collection efficiencies. Initially, calculations were time consuming, since each particle required a sample from the Poisson distribution. The time required for numerical simulation was greatly reduced with parallelization using CUDA GPU-based acceleration⁹¹. SEY at each stage was approximated by using the SEY curve; however, interstage collection efficiency was modified iteratively to achieve an average photomultiplier gain very close to that of the measured device. This method was deemed acceptable due to the highly variable nature of multiplier gain. Variability in secondary emitter growth leads to minor differences in SEY between PMTs and, indeed, dynodes of the same device. The exponential nature of electron multiplication in PMTs means that

similar devices operating at the same voltage may only achieve gains within the same order of magnitude. Table 3.2 shows the SEY and interstage collection parameters used to achieve the final result.

Table 3.2: *The yields and collection efficiencies used to numerically cascade the SER at D_6 to the anode.*

SEY	4.32	4.32	4.32	4.32	4.32	4.32	4.32
Interstage CE	0.932	0.938	0.934	0.933	0.936	0.93	0.99

Below, in Fig. 3.16, the SER distribution for 1931 photoelectrons from 2600 simulated releases are plotted against a measured SER distribution provided by Wright¹⁸. Following Wright’s measurements, responses below the 600,000 e^- noise floor were removed. A number

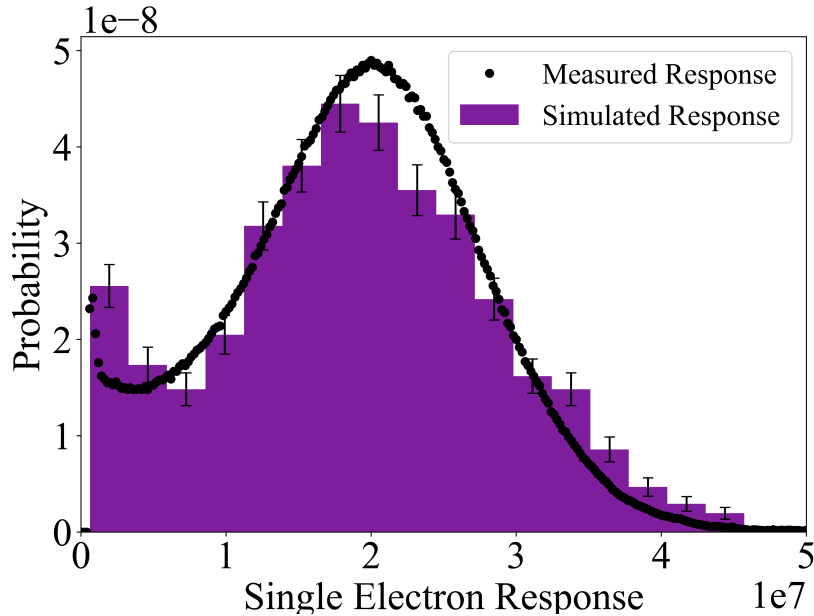


Figure 3.16: *The normalized distribution of 9214B photomultiplier gain from single photoelectron events are shown for experimental data¹⁸ (black) and for simulated data (purple). Uncertainty is $\sqrt{N}$.*

of performance metrics may be calculated from the SER which is measured using a low-intensity, gated photon source and has the advantage of being essentially noise free. The effective collection efficiency, η_{eff} , as described by Wright⁹², is the ratio of responses in the SER distribution to the number of photoelectrons released and cannot be directly measured,

as one cannot tell if a photoelectron was released and subsequently recaptured without a signal at the anode. Wright proposes a solution using a number of measurement techniques combined with simulation⁹²; however, η_{eff} may also be found directly through simulation as is done in this work. The single electron resolution is a measure of the variance in magnitude of primary signal pulses. This value may be presented as the standard deviation or FWHM of the predominate peak^{35;93}. The final performance characteristic that may be derived from the SER distribution is the ENF (Eq. 1.6), which is used to calculate the minimum multiplier contribution to a multi-electron signal such as those produced by a scintillation^{18;46;49}. The simulated and measured performance characteristics may be found in Table 3.3, below.

Table 3.3: *Simulated and measured 9214B electron transport results at 1250 V operating potential.*

	Simulated	Reported ^{18;92}	Error
Collection Efficiency	74 %	78 %	4.8%
Average Gain	$1.90 \times 10^7 e^-$	$1.88 \times 10^7 e^-$	+1%
Single Electron Resolution (FWHM)	$2.05 \times 10^7 e^-$	$1.81 \times 10^7 e^-$	+13.4%
Excess Noise Factor	1.25	1.21	+3.3%

Timing Timing characteristics for the 9214B PMT were simulated following the method described in 3.3.3. The electron collection rate at the anode for a single photoelectron release generally follows normal or skew-normal distribution due to transit time dispersion between the dynodes. This can be seen in the timing results of a full-transport and multiplication solution for the 9214B PMT shown in Fig. 3.17. Linear focus dynodes are well known for their compensating mechanism, which provides relatively tight timing dispersion. To find the timing characteristics produced via simulation, the number of electrons incident on the anode was collected in 0.25 ns intervals. Using R programming language, this distribution was differentiated to find the electron collection rate for each simulated photoelectron. A lower threshold of 25,000 total e^- was set to ensure 0.25 ns provided sufficient binning and low uncertainty. A skew-normal distribution was fit to electron collection rate, such that the mean transit time, rise time (10%-90%), and FWHM of the pulse could be calculated. Using the transit time distribution, TTS or “jitter” (FWHM) was also calculated. Distributions

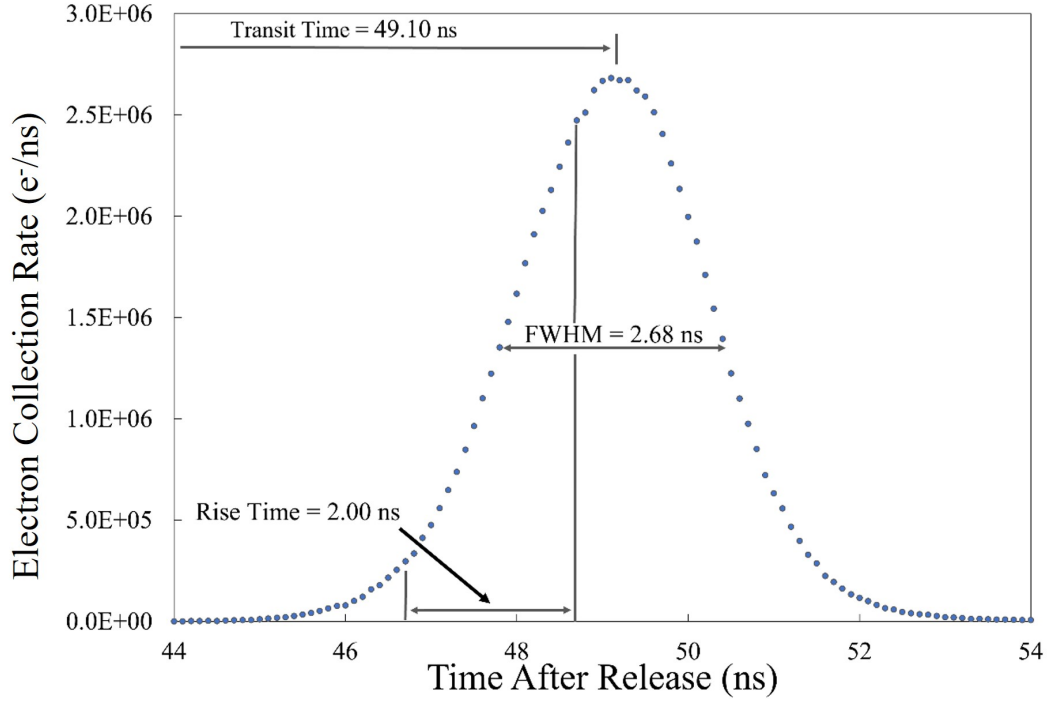


Figure 3.17: The electron collection rate at the anode for a simulated 9214B PMT. For this specific simulation, the cumulative number of electrons were recorded in 0.1 ns intervals. A number of timing characteristics are calculated as shown in the figure.

for each of these values are presented in Fig. 3.18, and the results are compared to nominal values provided by the manufacturer in Table 3.4

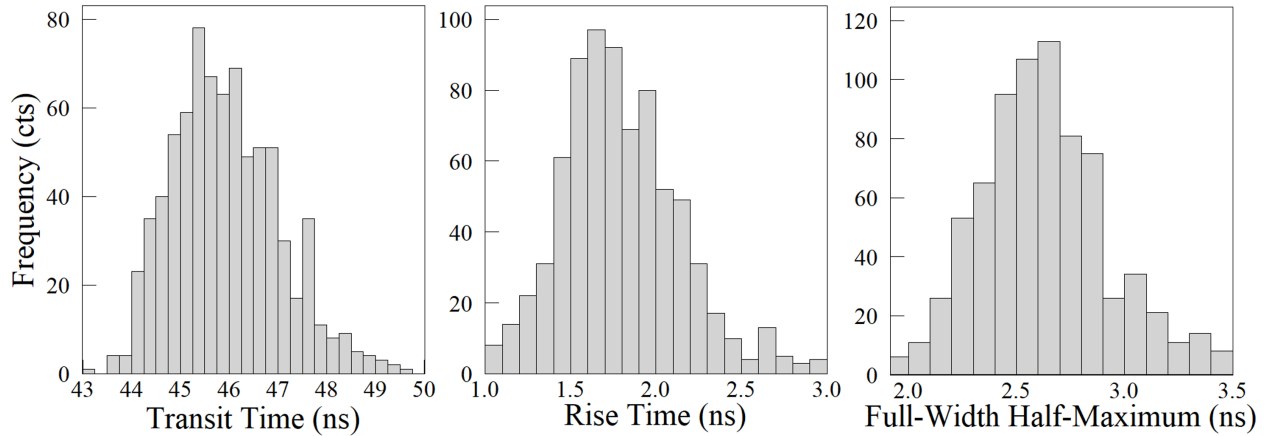


Figure 3.18: The timing responses for 770 pulses from 1300 simulated photoelectron releases across the full photocathode are presented. Transit time spread was found by analyzing the transit time distribution.

Table 3.4: *Single photoelectron timing performance at 1250 V operating potential.*

	Simulated	Reported ²⁴	Error
Transit Time	46.0 ns	45.0 ns	2.2%
TTS (FWHM)	2.6 ns	2.2 ns	18.2%
Rise Time	1.9 ns	2.0 ns	−5.0%
Pulse FWHM	2.7 ns	3.0 ns	−10.0%

3.3.5 Conclusions from Validation Model Results

SER results for the 9214B validation model, shown in Fig. 3.16, approximate the nominal values quite well, which is important for estimating the spectrometer energy resolution capability of novel devices. Gain is highly variable between devices, so a rough approximation beyond the sixth dynode was deemed appropriate¹⁸. Because of the chosen method of approximation, the simulated average gain is extremely close. With marginal modification to the recreated detector geometry, collection efficiency was within 5%, validating the particle transport method. Nonuniformity of the photocathode and dynode materials was not incorporated, meaning that not all contributions to the SER were considered¹⁸. This is less important for small dynodes; however, a PMT containing dynodes with an extended multiplication area may suffer from nonuniformity effects in the same way that separate PMTs of the same model do not have identical secondary emission properties¹⁸. Future work may incorporate random variance in the SEY across dynode surfaces to compensate; however, the author is not aware of any data quantifying such variance.

Timing results for the simulated 9214B (Fig. 3.18 and Table 3.4) also approximate the nominal measured values. Transit time and transit time spread are a measure of the full device’s timing performance, whereas rise time and pulse width better characterize dynode performance. Both the transit time and TTS overestimate nominal values; however, transit time is quite close. This suggests that the extraction field at the photocathode may not be perfectly representative of a typical device. Responses, shown in Fig. 3.18, with extended transit times (> 48 ns) may be attributed to photoelectron releases at the periphery of the photocathode where the electric field gradient is lower. Often, manufacturer’s will illuminate the inner 80% of the photocathode to reduce such edge-effects¹⁸. Additionally, for full, even

illumination, photocathode nonuniformity and reflective internal structures cause certain areas of the cathode to emit preferentially, which could tighten the transit time distribution. The pulse-width error is an acceptable -10% . Pulse width measures the dispersion of electrons as they transit the dynodes. Any fluctuation in dynode spacing and orientation will affect the electric field as well as emission trajectories and thus, the time dispersion.

Although the results of the validation model are promising, it is important to understand them contextually — to understand the limitations of simulation. Phenomenological models cannot capture every nuance of electron emission, nor can experimental measurements quantify every parameter. Because of the inherent limitations, it is best to consider gain a rough estimation. Excess noise factor, in part, dictates the relative variance and is largely a product of the first-stage SEY and collection efficiency, which are important for maintaining good spectrometer energy resolution. This value is less affected by exponential electron multiplication, and simulated values may be regarded as a decent approximation with some caveats regarding nonuniformity. Electron transport using FEM is quite accurate, provided a sufficient mesh. Overall, it is best to look at the broader scope (collection efficiencies, ENF, timing) with knowledge of typical device performance when using simulation as a tool to develop novel designs as is the case in subsequent sections.

3.4 “Catoptric” PMT Design and Simulation

The knowledge and confidence obtained by successfully simulating a well-characterized commercial PMT was applied to a new effort to design a novel rectangular PMT. The proposed device uses a reflective K_2CsSb photocathode deposited on a metal substrate. The reflective nature of the photocathode means that some emission properties, such as long-wavelength response and uniformity, will be inherently different than the validated head-on PMT model; however, no changes were made to the photoemissive properties of the photocathode. The dynodes, too, feature K_2CsSb as the secondary emitter. The SEY and backscatter coefficient at normal incidence have been altered to model the bialkali material; however, the energy distributions of the three emission modes have not been altered. Similarly, the affects of

incidence angle on the SEY and backscatter coefficient are consistent with the validation model.

3.4.1 Catoptric PMT Design

An iterative design process produced a 51.8×152.4 mm² rectangular window PMT having a miniaturized cross-section, extended three-stage multiplier and anode. Figure 3.19 shows a labeled section view of the virtual assembly. A curved photocathode improves photoelectron

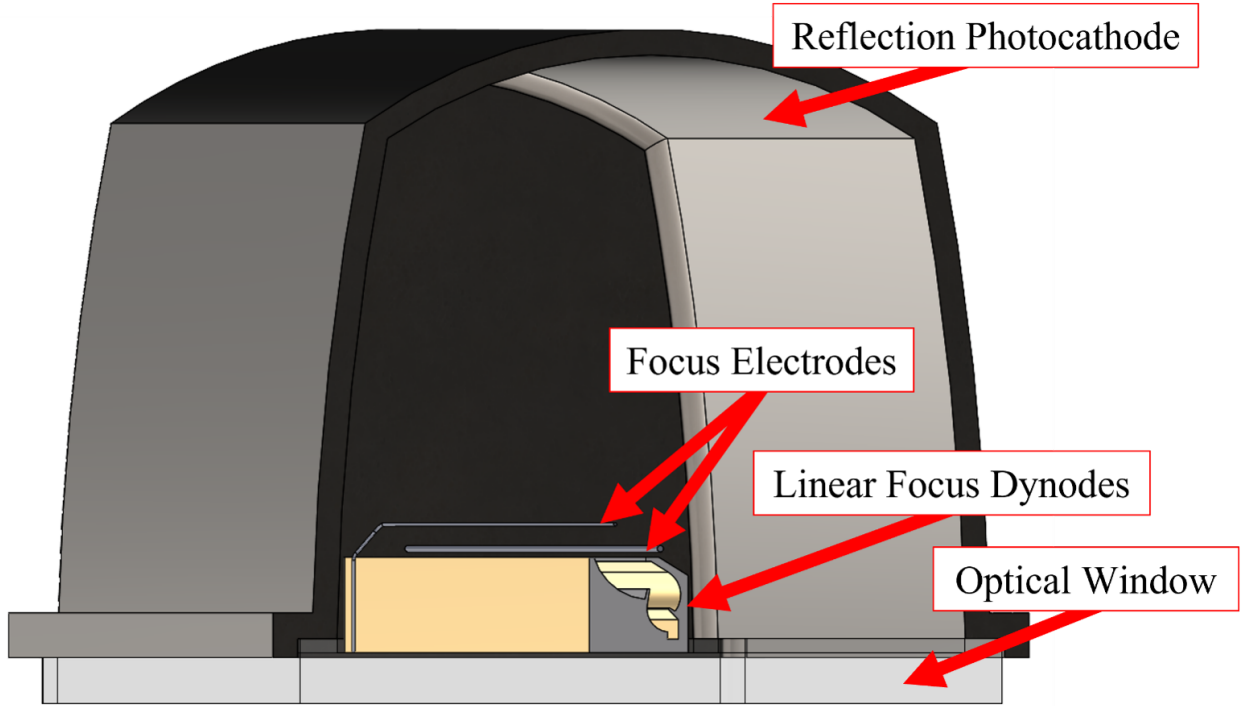


Figure 3.19: A 3D model of the simulated Catoptric PMT assembly. Gold-colored surfaces on the dynode structure highlight metalized areas which serve as the focus-electrode and sensitive dynodes.

collection efficiency, while reducing the transit time difference for photoelectrons released at the edges. The metal envelope, emulating a Winston cone, in conjunction with two small focusing electrodes serve to guide photons to the photocathode and photoelectrons to the first dynode stage. The general dynode shape was modeled after standard linear focus dynodes and does not require internal focusing electrodes to improve interstage collection efficiency. The high extraction field quickly guides secondary electrons to the subsequent stage, and

the design also provides some time-compensation which reduces the transit-time spread and pulse widths. Table 3.5 describes the standard voltage distribution used for simulation. The

Table 3.5: *A typical voltage distribution used for Catoptric PMT simulations.*

K	D ₁	D ₂	D ₃	A
2R	R	R	R	R

geometry of the simulated Catoptric PMT has fewer components than a typical PMT, which simplifies assembly, in theory. The dynodes have a wide range of feature sizes and a large aspect ratio, meaning that the final structure must be fabricated in one or two solid parts to mitigate tolerance-stacking issues that may otherwise affect electron transport.

3.4.2 Catoptric PMT Simulation

The results of 750 V electrostatic and charged particle tracing simulations are shown in Fig. 3.20. The electrostatics surface plot illustrates the electric potential distribution, while the overlaid black lines indicate a 20 V/div. equipotential distribution within the vacuum. The large spacing between the photocathode and 20 V equipotential line is indicative of a relatively low electric field, and the effects can be seen through the charged particle tracing results, below.

Collection Efficiency and Gain Average gain and photoelectron collection efficiency measured over the entire photocathode area monotonically increases with higher operating potential. It is evident that higher electric fields near the photocathode more effectively guide photoelectrons towards the first dynode. Below, in Fig. 3.21, simulated gains and collection efficiencies are displayed for many operating potentials. To meet gain and power consumption targets, the device will likely be operated between 550 V and 900 V.

Catoptric PMT Simulated SER Using the advanced simulation techniques, SER has been tabulated for 2,500 unique photoelectron releases. These results, plotted in Fig. 3.22, were obtained at 750 V operating potential. A minimum response of 2 electrons was included.

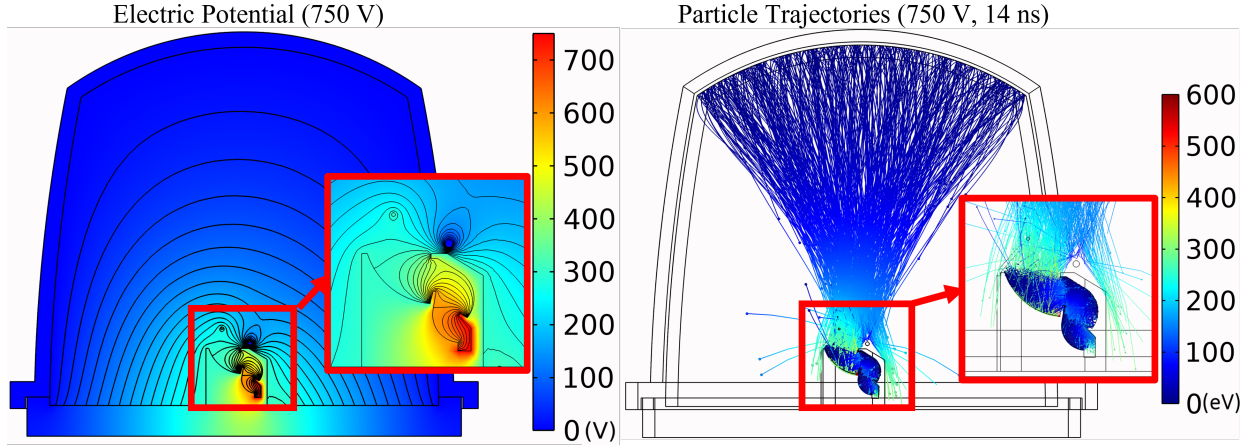


Figure 3.20: Left) Electrostatics simulation of the Catoptric PMT at 750 V operating potential plotted on the plane of symmetry. Right) Electron transport and multiplication at 750 V operating potential and 14 ns after release. Note that some photoelectrons released near the edges overshoot the first dynode.

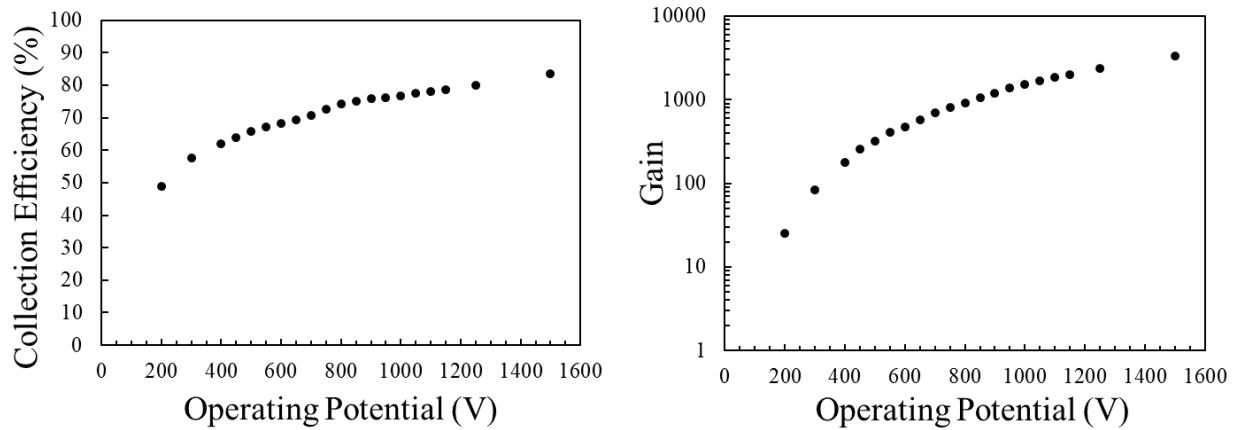


Figure 3.21: Average gain and photoelectron collection efficiency of the Catoptric PMT from 200 V to 1500 V operating potential for 1000 photoelectrons pseudorandomly distributed over the entire photocathode area.

A primary peak, mean of 1,437, represents the full PMT gain, whereas a collection of smaller pulses between 2-400 are products of backscatter and imperfect interstage dynode collection efficiency. The ENF for the Catoptric device is 1.29.

Collection Efficiency and Gain Uniformity Gain and photoelectron collection efficiency dependence on photoelectron release position were quantified over 100 regions of equal area across the photocathode. 100 simulations comprising the full area were performed at 750

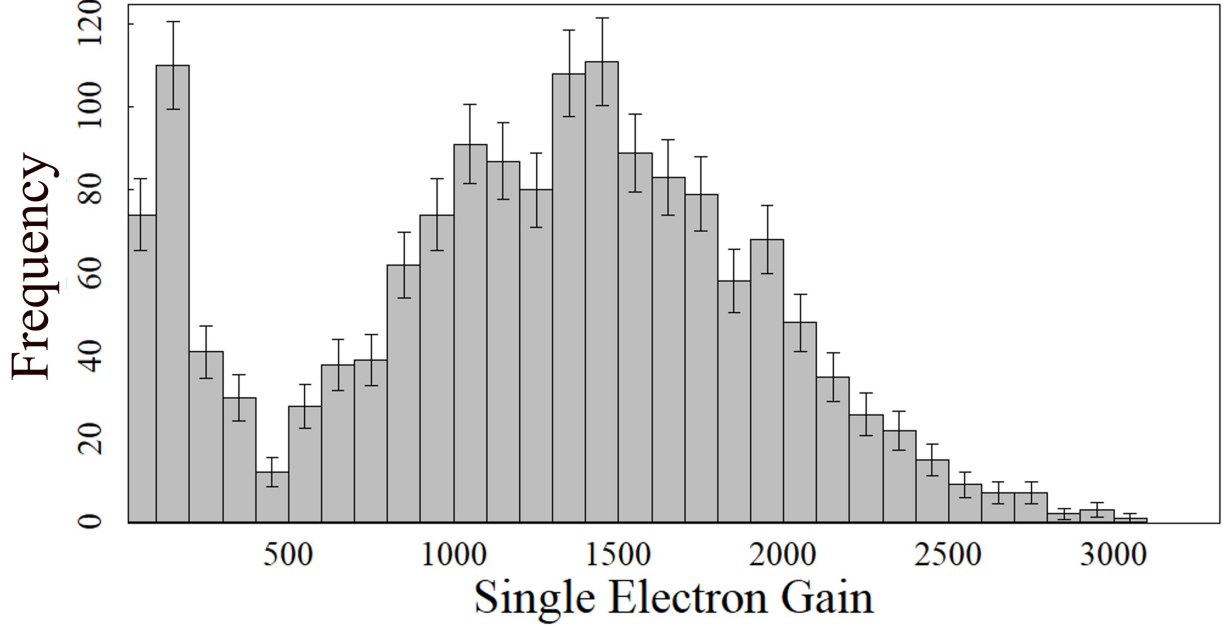


Figure 3.22: *SER for 2500 unique photoelectron releases at 750 V. Minimum included response is 2 electrons. A total of 1,537 responses meet this criteria meaning the effective collection efficiency at 750 V is $\eta_{eff} = 61.5\%$.*

V operating potential. 500 photoelectrons were released per simulation. Figure 3.23 illustrates the Catoptric device's response uniformity. Photoelectrons released near the sidewalls are not as effectively guided towards the first dynode. Additionally, photoelectrons released from the areas with the lowest performance approach the first dynode from the "back" side leading to overshoot, as seen in Fig. 3.20. Nearing the corners, both collection and gain are improved as the surrounding grounded substrate "pushes" photoelectrons on a favorable trajectory towards the center. Contrarily, approaching the midline near the endwall, the low electric field cannot as efficiently steer photoelectrons.

Catoptric PMT Simulated Timing Since the relatively low gain of the Catoptric device did not always form a well-defined peak with fine time resolution, a different method of simulating the transit time was employed. For each operating voltage, 50,000 photoelectrons were released from pseudorandom locations across the photocathode. SEY was reduced to 1, and each collected electron was regarded as the peak of a single electron pulse. The rate of electron collection at the anode was taken to be representative of the transit time distribution.

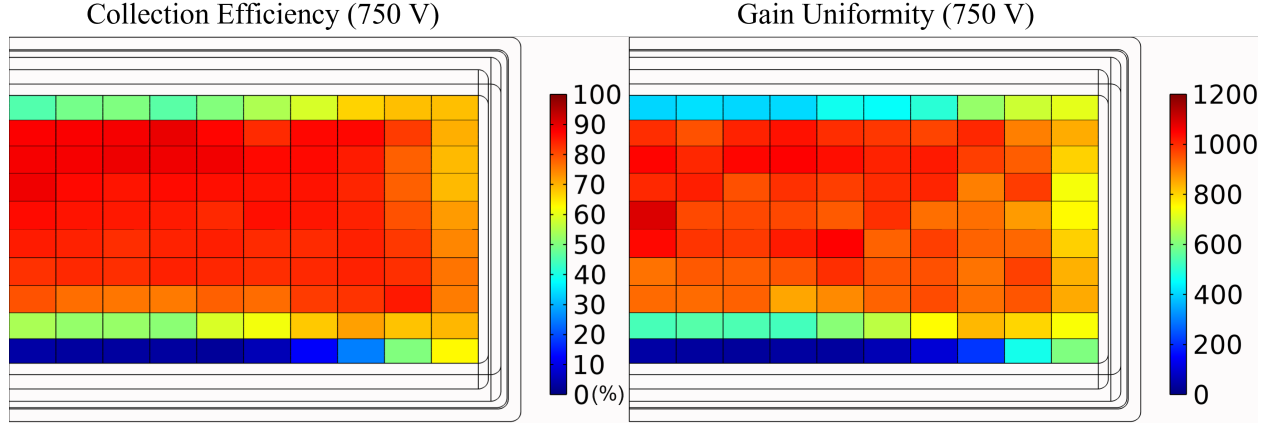


Figure 3.23: *Left) Spatial uniformity of photoelectron collection efficiency at the first dynode. Right) Spatial uniformity of gain. Heat map values correspond to equal-area regions from which photoelectrons were released.*

Electrons were collected at the anode in 0.1 ns intervals. Transit time distributions for various operating voltages are displayed in Fig. 3.24. Transit time (measured to the peak) and TTS are both reduced at higher K-D₁ voltages. Lower extraction fields near the photocathode periphery (Fig. 3.20) produce the longer transit times seen in the distribution. At 750 V operating potential, the peak transit time is 14.9 ns, while the TTS is 2.7 ns FWHM. Compared to typical 50.8 mm diameter, 8-10 stage linear focused devices, transit time is much faster due to the reduced number of stages, while TTS is not improved as a result of suboptimal front-end electron optics. Although relatively long tails are present in each of the distributions below 1500 V, the Catoptric device's timing characteristics are sufficient for fast inorganic scintillators, such as LaBr₃(Ce).

Table 3.6: *Simulated peak transit time and TTS for the Catoptric PMT at various operating voltages.*

Operating Potential	Transit Time (ns)	TTS FWHM (ns)
550 V	17.3	3.2
750 V	14.9	2.7
950 V	13.4	2.4
1150 V	12.2	2.1
1500 V	10.7	1.7

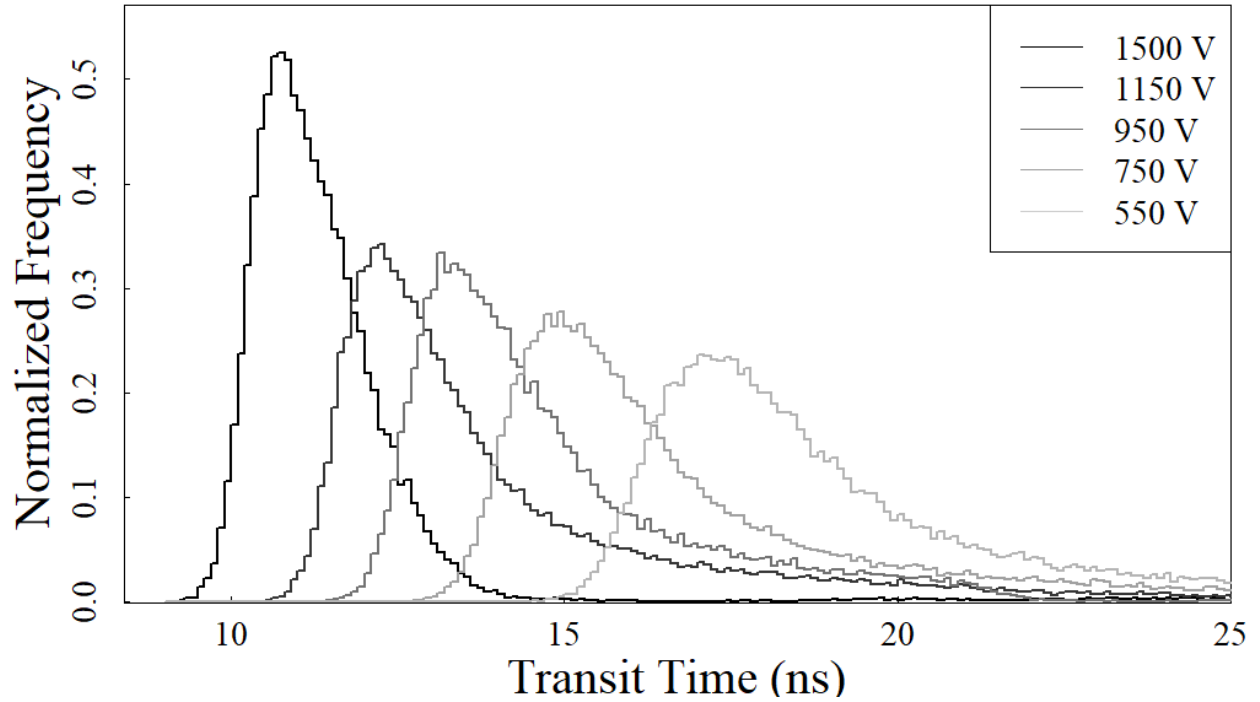


Figure 3.24: *Transit time distributions for single photoelectron pulses at the anode for various operating voltages. Higher electric fields near the photocathode at 1500 V greatly reduces transit times for photoelectrons released near the edges.*

3.4.3 Catoptric PMT Design and Performance Discussion

The new rectangular Catoptric PMT design utilizes a large-area optical interface to ensure efficient coupling to high-efficiency rectangular scintillators. Reflective sidewalls focus photons onto a smaller photocathode area to reduce QE and photoelectron collection nonuniformity. A metal-backed reflection-mode photocathode was chosen to increase QE. An additional benefit is that linearity is improved in high light-flux applications¹⁶. QE may be further enhanced through optical interference techniques by tuning the thin film thickness and reflective layers⁹⁴.

Offsetting the photoemissive material from the scintillator may also improve resolution in large scintillators by more evenly distributing light over the entire photocathode, thereby reducing the effects of QE and anode response nonuniformities^{18;73}. A miniaturized linear-focused dynode set with three multiplication stages and an anode was developed to minimize light obstruction at the optical interface. To accommodate the reduced gain, the Catoptric

PMT will take advantage of relatively large scintillation photon yields while leveraging sensitive, high-gain electronics to recover the signal magnitude.

A standard 50 mm PMT used for gamma-ray spectroscopy, the 10-stage ET Enterprises 9266B, has a relatively slow transit time of 40 ns compared to the Catoptric device transit time of 14.9 ns. The Catoptric PMT also exhibits a tighter TTS of 2.7 ns versus 6.5 ns for the 9266B. Still, the miniaturized dynodes and large, photocathode area inhibit faster photoelectron transport in the front-end, which is the primary contributor to TTS. The 9266B has a nominal gain of 6×10^5 at 850 V, while the Catoptric device is estimated to produce 805 electrons per photoelectron at 750 V on average, owing to fewer multiplication stages⁹⁵. ET Enterprises reports a “good” SER for the 9266B, meaning the ENF is ≈ 1.2 , whereas the Catoptric device simulations provide an ENF of 1.29. Standard box-and-grid dynodes, like those in the 9266B typically have a photoelectron collection efficiency greater than 90%¹⁶ at the first dynode, while the Catoptric device displays a first-dynode CE of 72%. Due to the Catoptric device’s relatively high ENF and low CE, the photon-collection, response uniformity, and QE characteristics are imperative to ensuring good spectroscopic performance.

3.5 Transmissive PMT Design and Simulation

Additional transmissive PMT design studies resulted in a geometry more closely resembling a standard “head-on” PMT. A transmissive photocathode emits photoelectrons in the direction of incident photons. Photoelectrons are then guided to the first dynode. Secondary electrons cascade through subsequent dynodes until reaching the collection anode. The transmissive design, shown in Fig. 3.25, provides a number of advantages that are immediately apparent. Namely, larger, more effective focusing grids and dynodes with higher interstage collection efficiencies. Dynode and electrode components using common sheet-metal forming and spot-welding techniques may reduce fabrication difficulty and cost. An open volume behind the dynode structure offers a large region for vacuum and electrical feedthroughs, if needed. Conversely, a number of disadvantages also exist when comparing the transmissive

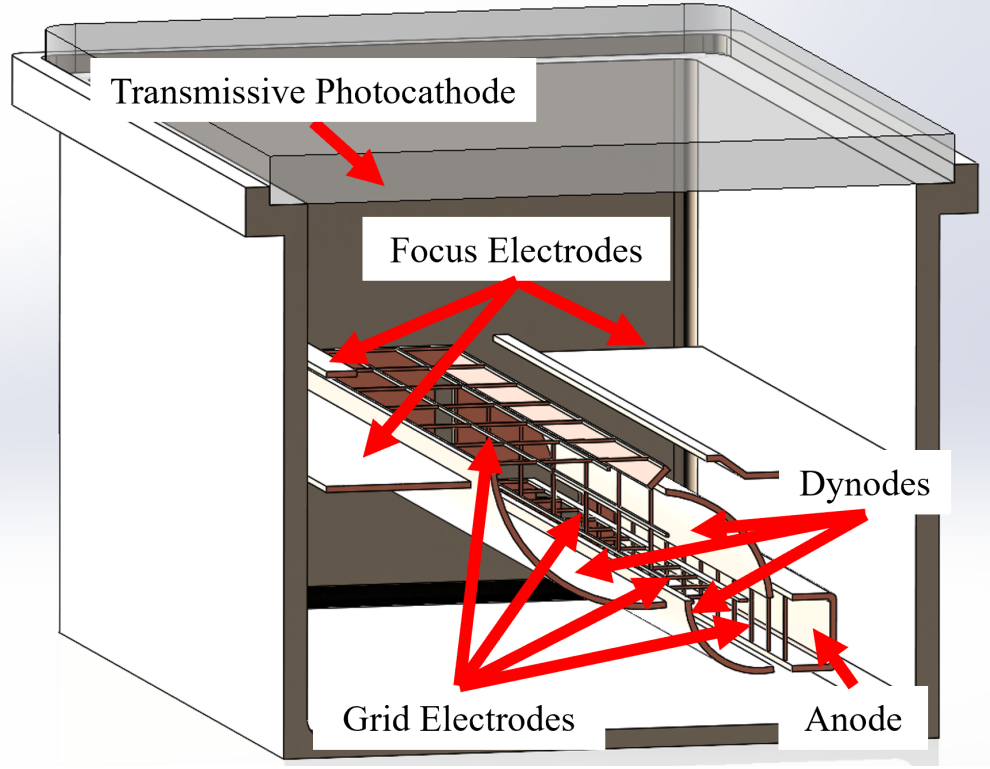


Figure 3.25: *A section view of the simulated transmissive PMT assembly with large box-and-grid dynodes.*

PMT to the Catoptric design. The close proximity of the photocathode to the scintillator means that photons emitted near the window will be tightly distributed and the signal will be highly dependent on the local anode response, as discussed in Sec. 2.2. Moreover, standard transmissive photocathodes are thin, making them less efficient with regards to photoconversion. Striking a balance between photon and photoelectron absorption lengths typically produces semitransparent cathodes with QEs on the order of 20%-25%. Modern growth techniques have produced transmissive photocathodes with QEs as high as 45% by tightly controlling the film morphology^{78;96;97}.

3.5.1 Transmissive PMT Simulation Results

An initial design/simulation effort has yielded a conceptual transmissive device of similar size to the proposed Catoptric PMT. A $50.8 \times 152.4 \text{ mm}^2$ photocathode is mated to a metal housing held at ground potential. Ceramic endplates separate the housing from the focus

and multiplier electrodes which use the same voltage distribution as the Catoptric device (Table 3.5) and may be seen in Fig. 3.26.

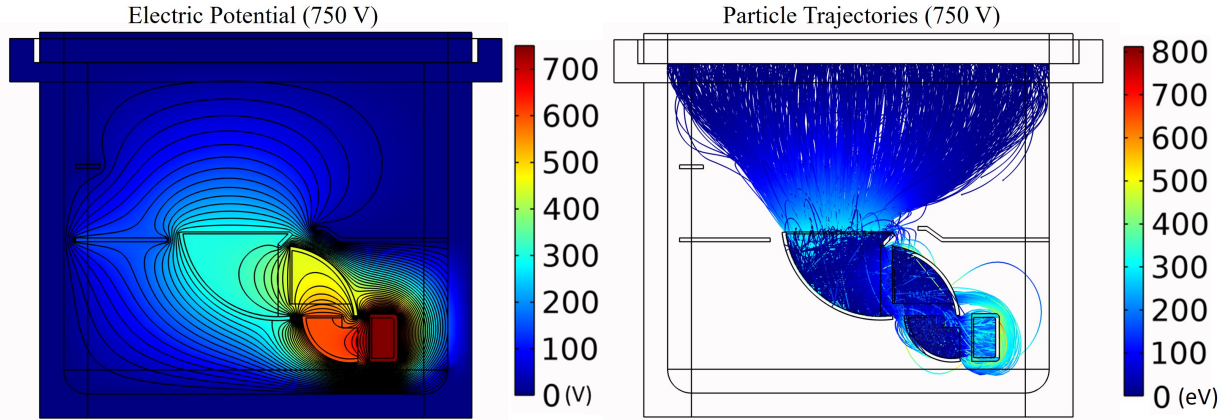


Figure 3.26: Left) *Electrostatics simulation of the Catoptric PMT at 750 V operating potential plotted on the plane of symmetry.* Right) *Electron transport and multiplication at 750 V operating potential and 25 ns after release.*

The initial transmissive PMT iterative-design effort did not produce a viable linear-focused dynode design due to their relative complexity. The standard linear focus dynode cross-section consists of a linear segment mated tangentially to a partial circular-arc. An array of these dynodes extends away from the photocathode. Another set are mirrored across the multiplier’s mid-plane and are translated some distance along the PMT axis midway between the opposing electrodes. No intermediate focus-electrodes are used to enhance electron collection or extraction. Instead, the electric field within the multiplier structure is purely the result of the dynode geometries (refer to Fig. 1.22). Therefore, the length and angle of the linear segment, the radius and extent of the arc segment, and the axial and transverse spacing between dynodes are parameters imperative to ensuring a uniform electric field across the dynode surfaces. As stated in Sec. 1.5.6, optimized linear focus dynode designs provide uniform, high extraction field gradients, good timing, and efficient secondary electron collection; however, the large parameter-space makes converging to a functional design much more difficult. Additionally, transitioning from the first dynode stage — optimized for photoelectron collection — can be quite challenging.

A box-and-grid multiplier design was chosen instead, with the dynode “box” cross-section

being 90° circular arcs of similar radius. Grid electrodes simplify secondary electron transport optimization compared to the much more complex electric field optimization required for linear focus multipliers. Below are the results of the preliminary transmissive PMT design study.

Collection Efficiency and Gain Average gain and photoelectron collection efficiency at the first dynode were simulated at multiple operating potentials. 1000 photoelectrons were released spanning the entirety of the photocathode area. The results are plotted against operating voltage in Fig. 3.27, below. Similar to the Catoptric device, average gain monotonically increases across the simulated potentials; however, collection plateaus at 650 V. Both collection and gain are significantly higher across all operating potentials for the transmissive device — a consequence of larger collection surfaces.

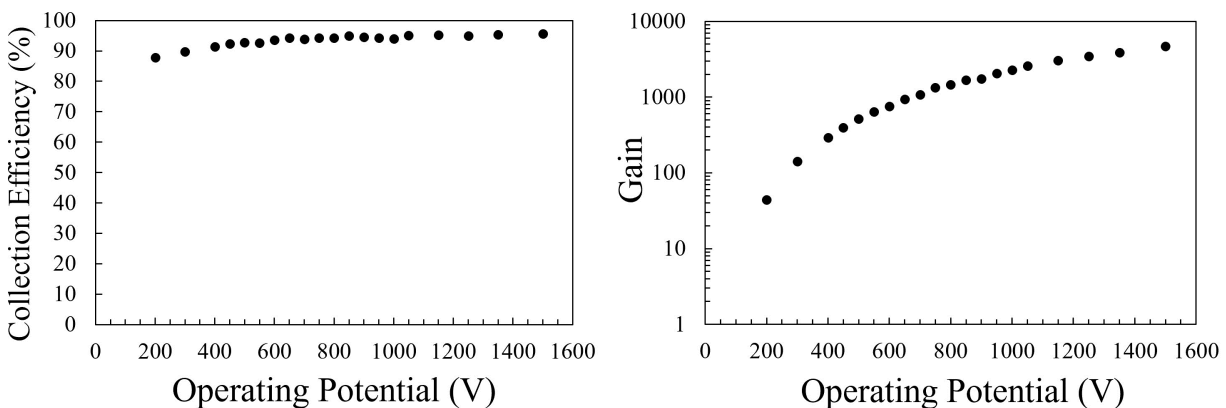


Figure 3.27: *Average gain and photoelectron collection efficiency of the Transmissive PMT design from 200 V to 1500 V operating potential for photoelectrons randomly distributed over the entire photocathode area.*

Transmissive PMT Simulated SER Figure 3.28 illustrates the transmissive SER produced by 3,375 unique photoelectron releases evenly distributed across the photocathode. Again, the SER is composed of two primary peaks, with undersized signals produced by backscatter and imperfect collection efficiency. The percentage of undersized signals is noticeably smaller for the transmissive design given the larger effective area of the dynodes. Additionally, the full-signal peak's mean SER is much larger, at approximately 1830 elec-

trons per photoelectron. This is a product of more favorable interstage electron trajectories. The transmissive PMT has a simulated ENF of 1.30.

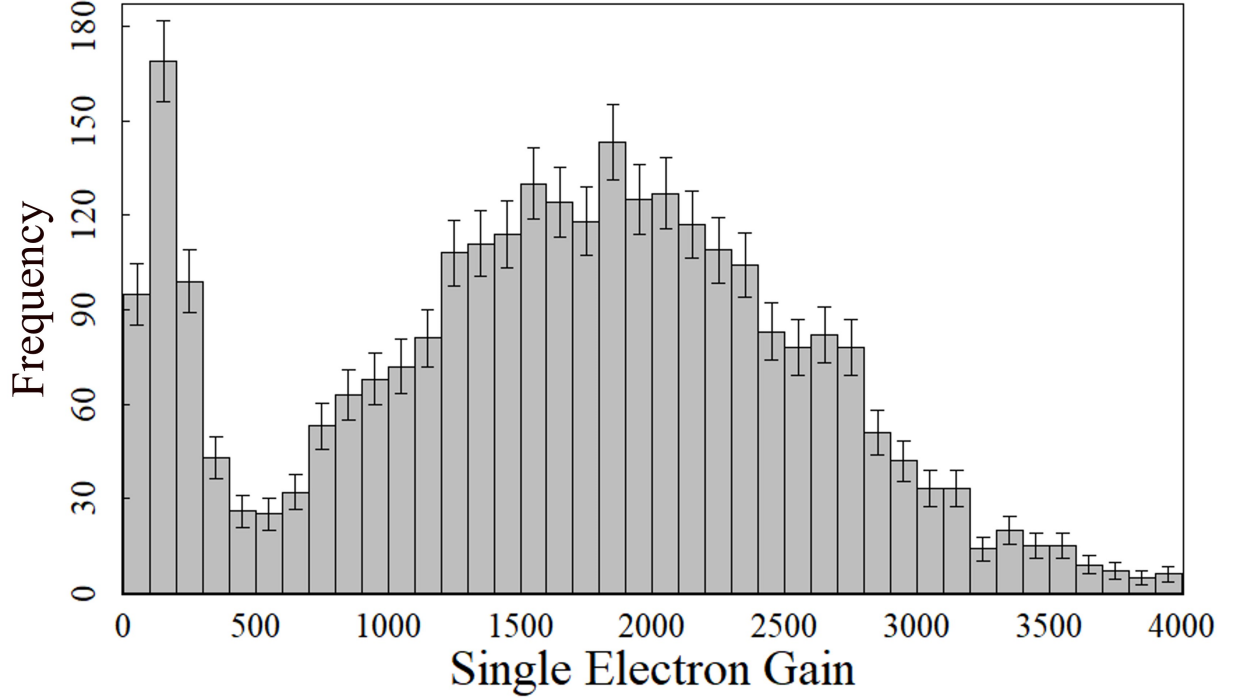


Figure 3.28: *SER of the transmissive PMT simulations for 3,375 unique photoelectron releases at 750 V. Minimum included response is 2 electrons. 2,836 responses are larger than the minimum response, meaning $\eta_{eff} = 84\%$ — far exceeding that of the Catoptric device.*

Collection Efficiency and Gain Uniformity Spatial gain and photoelectron collection efficiency uniformity were simulated over half of the transmissive photocathode. The cathode was divided into 150 equally sized regions, over which 500 photoelectrons were emitted per release. For these studies, electron transport was again simulated at 750 V operating potential. Both gain and collection magnitude and uniformity are greatly improved compared to the Catoptric device and can be seen in Fig. 3.29. Photoelectron collection again suffers when released from the back side of the first dynode, as some photoelectrons overshoot the target. However, gain is not degraded to the same degree given the larger effective area of D_1 .

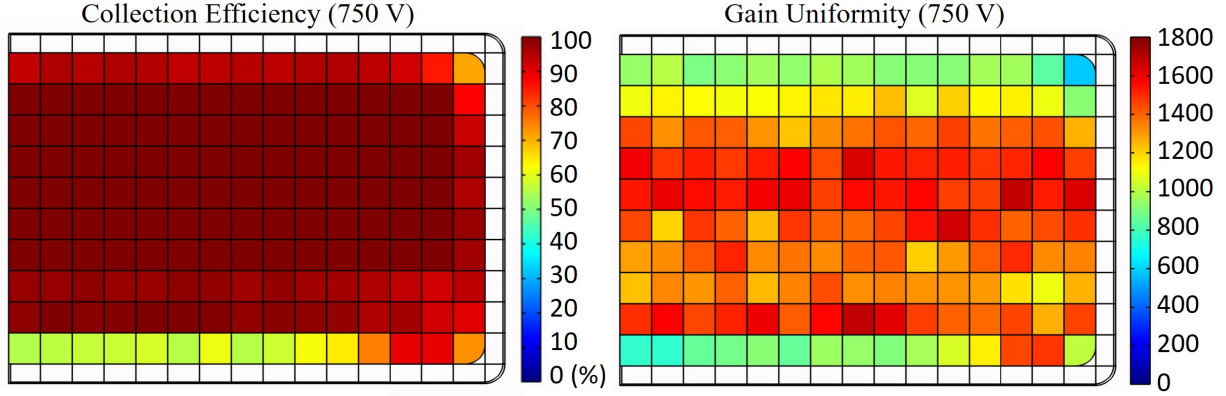


Figure 3.29: *Left) Spatial uniformity of photoelectron collection efficiency at the first dynode. Right) Spatial uniformity of gain. Heat map values correspond to equal-area regions from which photoelectrons were released.*

Transmissive PMT Simulated Timing The transmissive PMT transit time distribution was simulated following the Catoptric simulation method. 50,000 photoelectrons were released from pseudorandom locations across the photocathode for each operating voltage. The transit time distributions at multiple operating voltages can be seen in Fig. 3.30. Transit time and TTS are both reduced at higher K-D₁ voltages, as expected. The extended tail for each distribution is clearly attributable to the low extraction field present on the “right” side of the transmissive PMT and at the dynode surfaces (Fig. 3.26). Additional efforts to design a symmetrical electric field for the front end would greatly reduce the number of lagging pulses. It is likely that some delayed pulses would persist due to edge effects; however, a plano-concave cylindrical lens could be incorporated as the optical window to improve timing similar to the plano-concave window of the 9214B. At 750 V operating potential, the peak transit time is 17.7 ns, while the TTS is 5.7 ns FWHM. Compared to typical 50.8 mm diameter, 8-10 stage linear focused devices, transit time is quite fast due to the reduced number of stages. However, the TTS is slower due to edge effects and unoptimized focusing electrodes. Table 3.7 shows the timing properties derived from the transit time distributions provided.

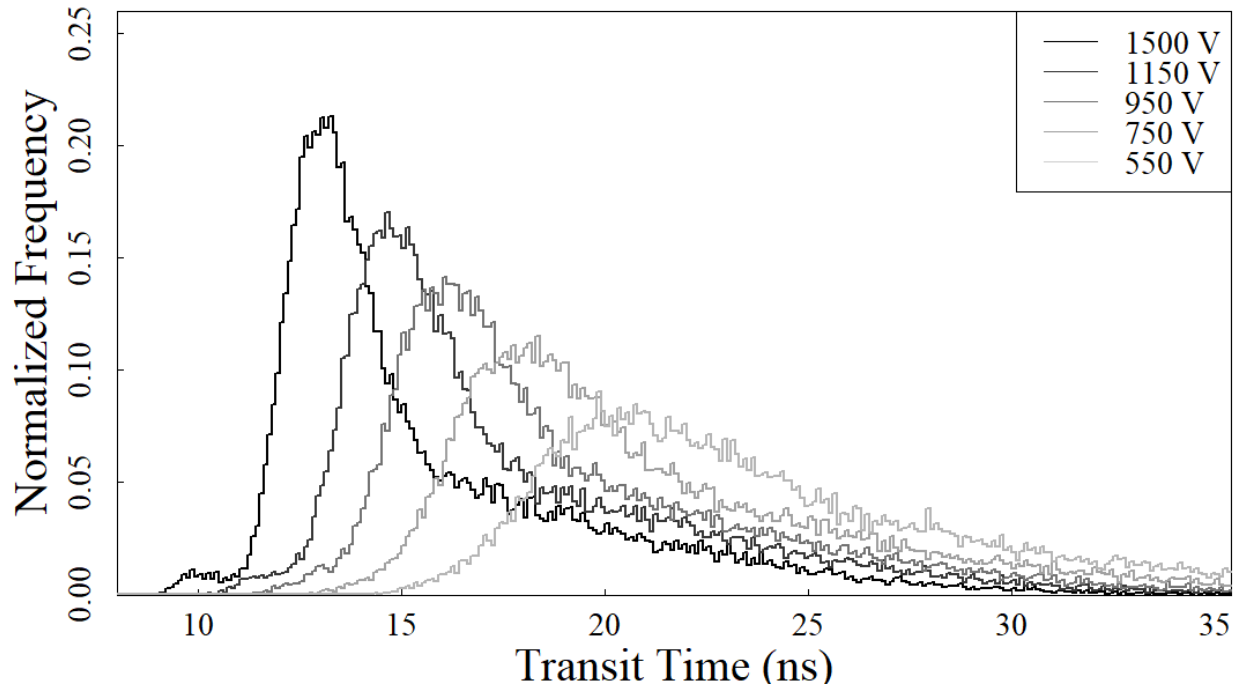


Figure 3.30: *Transmissive design transit-time distributions for single photoelectron pulses at the anode for various operating voltages. Box-and-grid style dynodes greatly extend the electron collection time at all potentials compared to their linear-focused counterparts.*

Table 3.7: *The simulated peak transit time and TTS for the transmissive PMT at various operating voltages.*

Operating Potential	Transit Time (ns)	TTS FWHM (ns)
550 V	20.3	7.7
750 V	17.7	5.7
950 V	15.9	4.1
1150 V	14.6	3.1
1500 V	12.9	2.7

3.6 Conclusions

A validated electron emission and transport model has been used to develop two novel PMT designs of similar size. The earliest and largest efforts focused on the design and optimization of the reflective photocathode PMT, since, in theory, reflective photocathode growth is simpler and may provide superior performance. Preliminary optical transport simulations suggest that the miniaturized dynode set obstructs just 10% of the transmitted photons; however, the drastic changes in refractive index as a photon travels from the scintillator,

through the optical interface, then vacuum, and finally, the photocathode will impart substantial reflective losses. Additionally, electron transport simulations have shown focus and dynode assembly positioning to be absolutely critical. A misalignment of <0.5 mm would drastically affect photoelectron collection and multiplication, meaning the performance of the photocathode is absolutely critical to the viability of the reflective design as a gamma-ray spectrometer. The fabrication process will be discussed further in subsequent chapters.

The proposed transmissive device mirrors more traditional head-on PMT designs. Both η_{eff} and gain are superior to the Catoptric design. Further development of the focusing grid and dynodes is required to improve timing and excess noise characteristics. Without multiplication stages obstructing the optical-transport region, both grids and dynodes may span larger areas. Therefore, the entire catalogue of dynode designs (Fig. 1.21) is available. A diverse range of PMT sizes may be more easily designed, and vertically-compact devices with enhanced response to magnetic fields may be produced using mesh or metal-channel style dynodes. The primary deficits of the transmissive device are potential photocathode nonuniformity, variable wavelength response, and lower QE, although recent work from the University of Chicago and Incom, Inc. have shown that high quality transmissive alkali photocathodes may be produced over a large area which is promising for this application³².

Both rectangular PMT designs offer a massive size advantage when compared to spectrometers using a commercial 127 mm diameter device, as seen in Fig. 3.31. The circular-window PMT does not cover the full crystal width, yet it has excess depth and height compared to the Catoptric or transmissive-style rectangular PMTs. Reducing the overall PMT size increases spectrometer mobility and modularity, while optimizing the optical interface will reduce spectrometer energy resolution by increasing the transfer efficiency.



Figure 3.31: *Size comparison of spectrometers using $50.8 \times 152.4 \times 152.4$ mm scintillator crystals. On the left, a 127 mm PMT is shown, while a model of the Catoptric PMT paired to the crystal housing is displayed on the right.*

Chapter 4

Catoptric PMT Component Fabrication

Following the initial Catoptric PMT simulation and design efforts, work quickly began on modeling and fabricating the required componentry. Conversations with researchers developing large-area MCP PMTs for high-energy physics applications guided the fabrication process outline⁵⁴. Component design and fabrication is discussed, below.

4.1 Envelope

The photocathode envelopes were produced in a three-step process. First, billet 304 stainless steel was machined on a 5-axis mill to the desired shape. The machined envelope is quite rough, so manual polishing was performed to bring the surface finish to a 0.38 μm Roughness Average (R_a). The tight, irregular shape of the envelope interior produced a nonuniform finish on both the photocathode and sidewalls. Sharp peaks left by the manual polishing process increase local electric fields at the photocathode which spreads photoelectron emission trajectories. Additionally, a mirror finish is preferred for the reflective sidewalls to promote favorable photon trajectories. A final electropolishing process was used to ablate peaks left by the mechanical processes, bringing the surface to a near mirror finish.

To complete the envelope fabrication, a grounded focus wire was laser welded into position, as were mechanical fixtures used to hold the dynodes in position. The final assembly can be seen in Fig. 4.1.

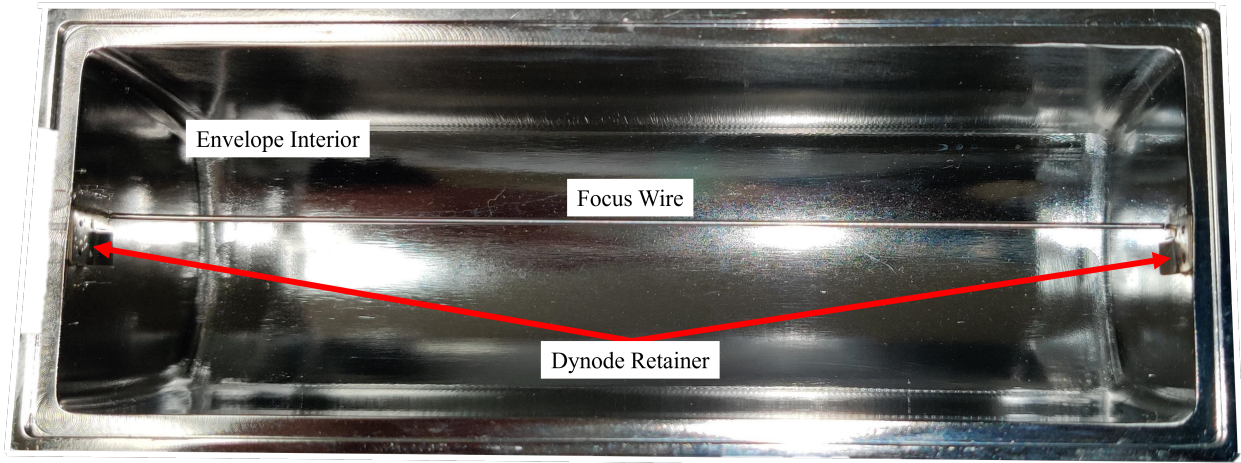


Figure 4.1: *The PMT envelope assembly with the vacuum housing, laser-welded dynode retainers, and the grounded focus wire.*

4.2 Window

A two-part assembly joins a machined 4 mm borosilicate optical window to a 1.1 mm frame of the same material using a non-conductive, hermetic glass frit seal. A 200 nm NiCr layer was sputtered onto the top-side of the thin frame. An additional 200 nm layer of Cu was sputtered atop the NiCr without breaking the Ar atmosphere to prevent contamination of the underlayer. Metalization serves to promote adhesion to the indium diffusion seal, which mates the window to the PMT envelope. Prior to frit-sealing the glass frame to the window, Cr/Ni, 30 nm/100 nm, electrical traces were electron-beam evaporated onto the window. An electroless nickel immersion gold (ENIG) metal deposition was then performed on the metal traces to increase thickness. The metal traces, in conjunction with the frit seal, should provide a hermetically-sealed electrical feedthrough to provide voltage and pass signal from the dynodes. Figure 4.2 depicts the metalized glass substrates.

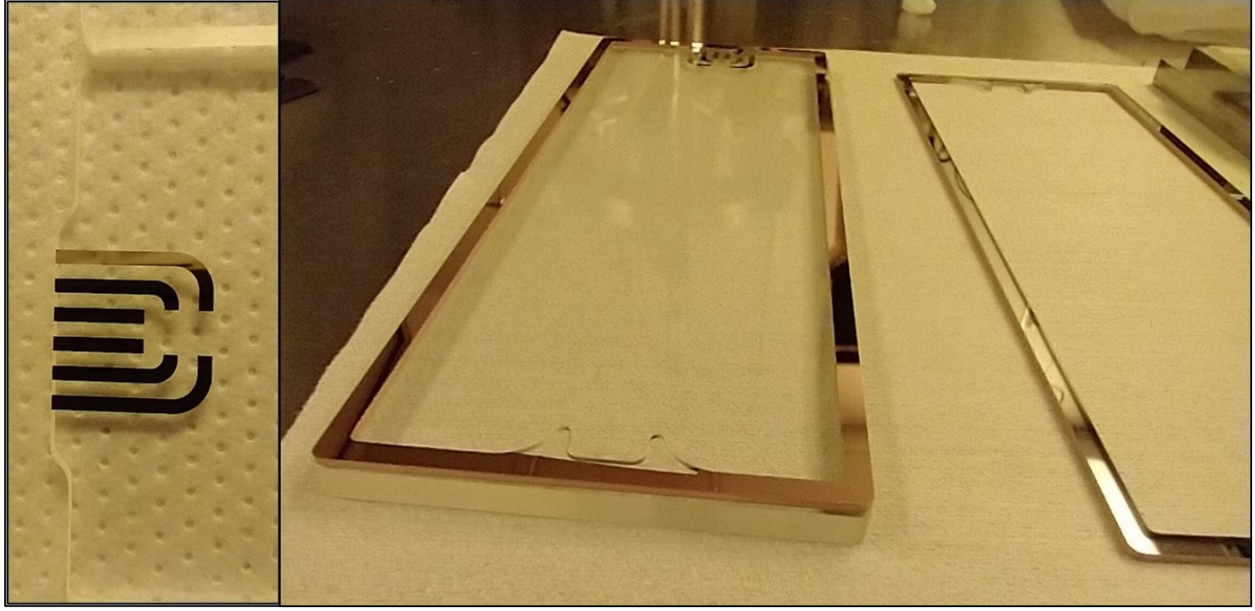


Figure 4.2: *Left) The metal traces which provide the electrical connection to each of the separate dynode electrodes. Right) A metalized frame sitting atop a metalized window. To the right, rests an additional metalized glass frame.*

4.2.1 Frit Seal

To create the aforementioned electrical feedthrough, the two glass substrates are joined using lead-free AP4115AB glass frit from AGC America⁹⁸. Frit seals have the distinct advantage of being very adaptable to rough or uneven surfaces, meaning the seals form even with the disruption of electrical contacts. AP4115AB is a $\text{Bi}_2\text{O}_3/\text{ZnO}$ frit made for $< 150 \mu\text{m}$ joints on borosilicate substrates. The frit is composed of ceramic and glass nanoparticles with CTE-modifiers to match the substrate and avoid cracking while forming the bond. Organic solvents and binders suspend these materials in a paste which may be ink-jetted or screenprinted onto a substrate as in Fig. 4.3. For screen printing, it is recommended that the mesh be greater than three times the mean particle size⁹⁹. The applied thickness is determined by the mesh size and may be chosen somewhat arbitrarily so long as any unevenness is sufficiently covered by the frit paste.

A 3 mm border for $155.6 \times 158.8 \text{ mm}^2$ windows, as well as another for $25.4 \times 25.4 \text{ mm}^2$ test coupons, was patterned on a 355 threads per inch ($34 \mu\text{m}$ wire diameter) mesh used for screenprinting. The frit was applied to both substrates using a screen printer, seen in

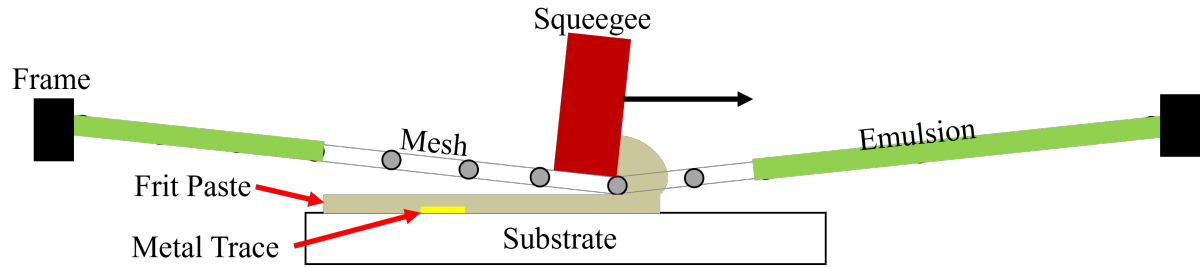


Figure 4.3: *A cross-sectional view of frit being applied to a metalized substrate.*

Fig. 4.4. The frit paste was then subjected to drying and burnout heat cycles (Fig. 4.5)

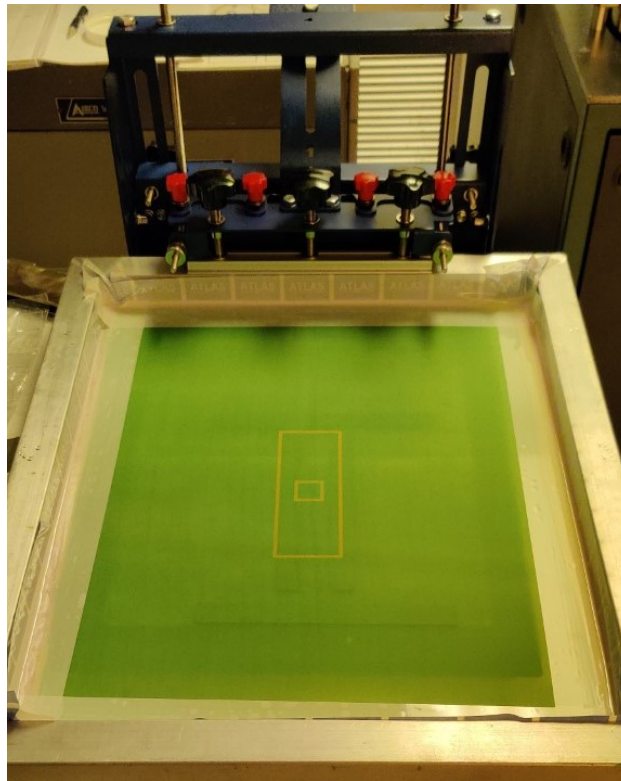


Figure 4.4: *Frit screen printing is performed with standard equipment and patterning process. Some consideration for compatible cleaning solvents must be made to avoid damaging the pattern.*

in an oven with a standard atmosphere and good airflow. Pressure was applied to the joint while cyclically translating the substrates in-plane a small distance to join the surfaces. The substrates were clamped in a fixture to apply pressure. A final glazing/bonding heat cycle was preformed in an air atmosphere to join the two parts. For the large bond area, a clamping

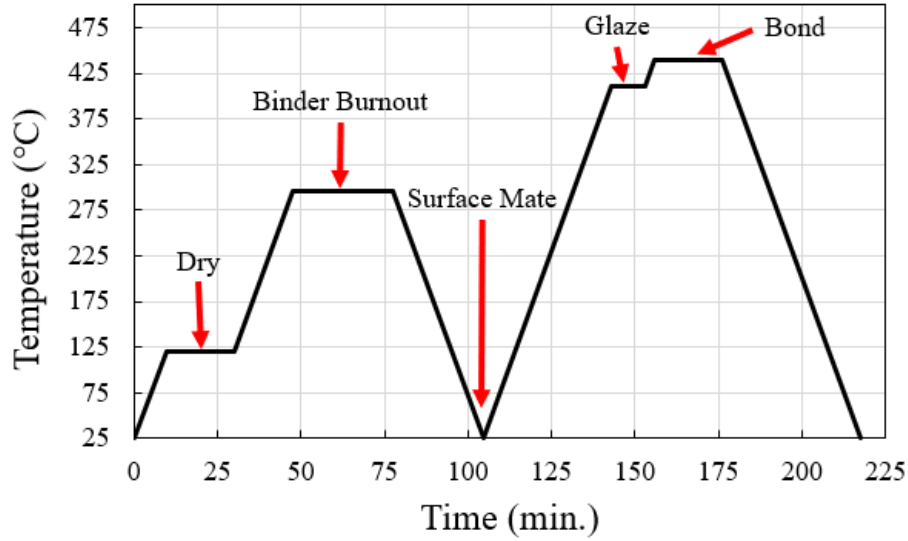


Figure 4.5: *The final temperature cycle used for frit bonding.*

force was necessary to promote surface wetting in the case of nonuniform thickness. Even still, small thickness nonuniformities in the dried paste ($<10\text{ }\mu\text{m}$) were sufficient to cause vacuum leaks in the final bond.

Dozens of trials were undertaken using test samples to parameterize the process and localize failure modes. Higher heat and longer exposure times were required at the various set-points to compensate for a measurable difference between the substrate temperature and that read by the oven. Figure 4.6 shows a number of failed bonds created under an N_2 atmosphere. Although the N_2 atmosphere was critical in maintaining the Cu metalized layer, it was eventually deemed unfeasible. Incomplete binder burnout, weak bonds, and partially oxidized Cu surfaces meant that the nitrogen-atmosphere bonding process could not produce a viable seal while maintaining an effective Cu layer for In bonding.

SEM images of a single-sided application reveal nonporous media with a bond thickness of approximately $65\text{ }\mu\text{m}$, and a contact angle of 10.5° , indicating good wetting of the coated surface¹⁰⁰. The final frit thickness for the dual-sided application was approximately $100\text{ }\mu\text{m}$. Frit-bonded coupons passing an initial visual inspection were placed in the tensile-strength testing apparatus shown in Fig. 4.8. A load cell was read out using a LabView program further described in 5.2.4. The $25.4\times 25.4\text{ mm}^2$ test coupons with 3 mm wide bonds

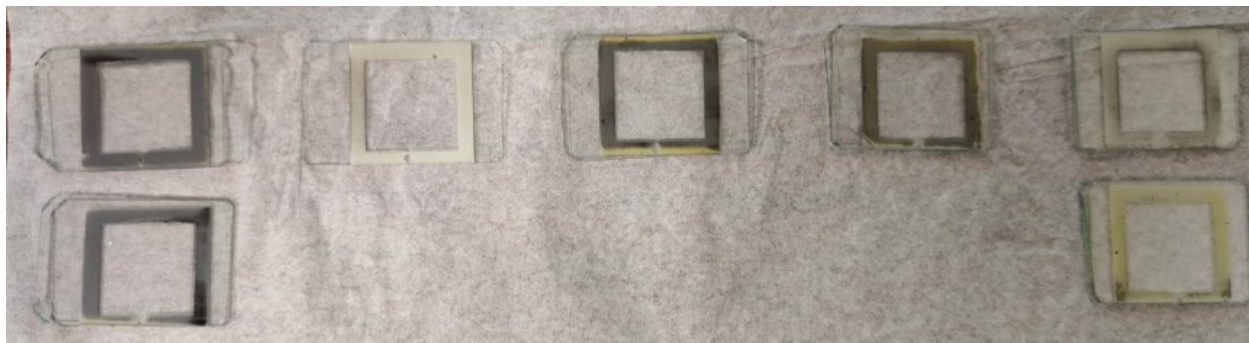


Figure 4.6: A number of weakly-bonded frit seals. Gray coloration indicates insufficient binder burnout as a result of the O_2 deficient atmosphere. The lighter colored seals failed due to premature firing.

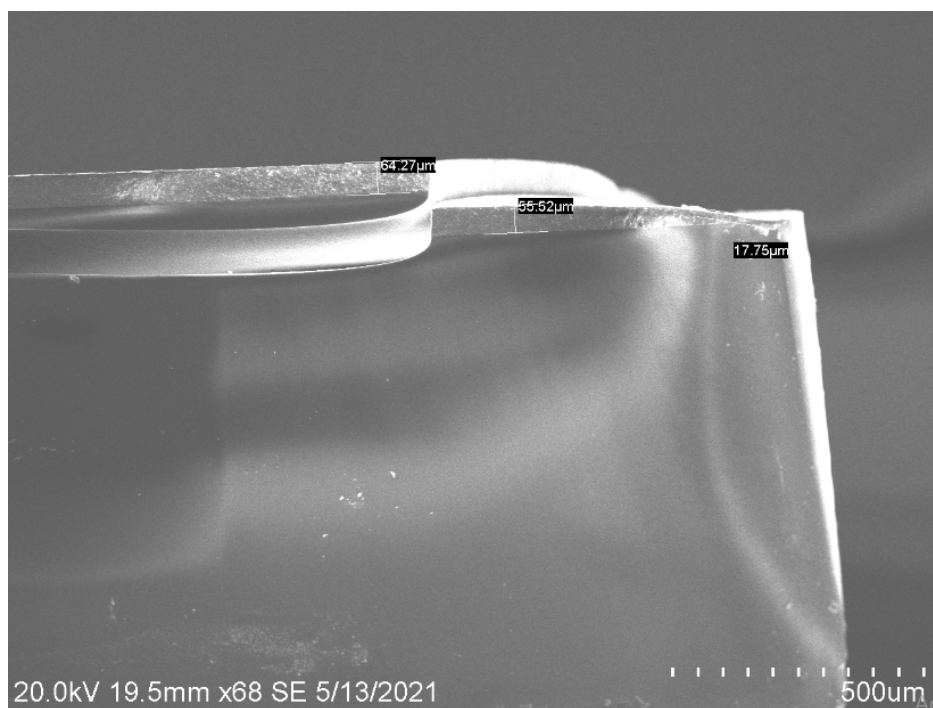


Figure 4.7: SEM image of a single-sided frit application. Courtesy Taylor Ochs, RDT.

were pulled until failure by tightening a nut at the top of the threaded rod. Successfully bonded samples had an average tensile strength of $280 \text{ kPa} \pm 23\%$. These tests served as a somewhat qualitative measure of bond characteristics. Improved tensile and shear strength quantification studies should use a standard universal testing machine with a high-accuracy load-cell and improved sample holding to reduce the shear induced by substrate flexure.

A custom hermeticity test adapter was fabricated by welding a flat plate to a KF16

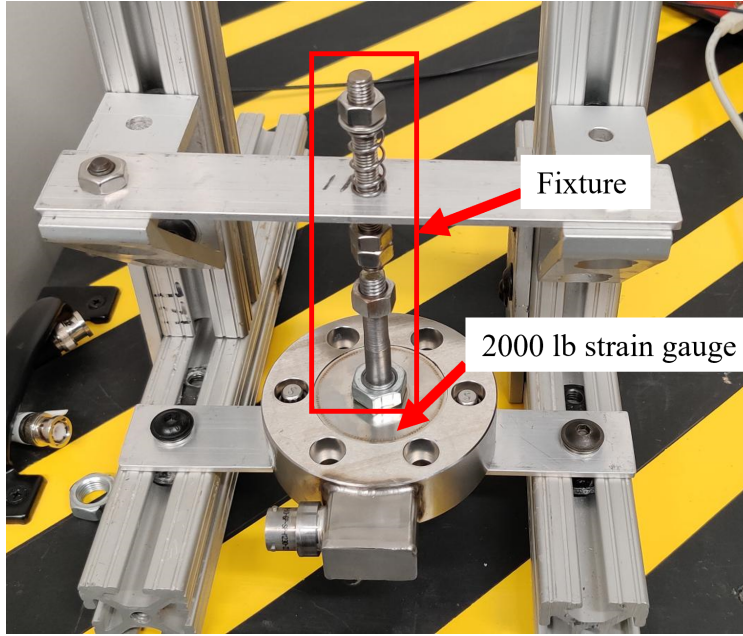


Figure 4.8: *The device used to test the tensile strength of frit-bonded coupons.*

flange. A Buna-N gasket was cut from a 3 mm thick sheet and used to seal glass assemblies to vacuum adapter. The frit-seal clamping fixture was also employed to clamp the vacuum test assembly together (Fig. 4.9). The KF16 flange was mated directly to an Oerlikon

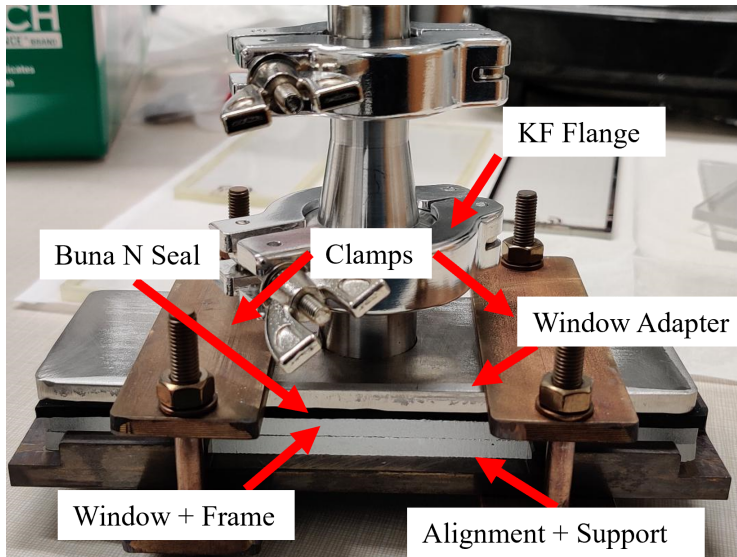


Figure 4.9: *The frit bond vacuum test assembly.*

Leybold PhoeniXL 300 helium leak detector with a 3.75×10^{-12} torr l/s detection limit. First, a flat piece of glass was tested to find the base leak rate of the gasket. The seal was

sufficient to exceed system capability. Several frit-sealed window assemblies and additional 4 mm thick surrogates were then placed in the test fixture. None of the 7 viable samples passed UHV testing, and the lowest leak rate observed was 5×10^{-5} torr l/s for one of the surrogates.

The particular frit paste used was purchased per the manufacturer's recommendation. Due to the difficulty of forming a seal, exploration of other frit compounds is recommended. A lower melt viscosity would likely negate the effects of coating nonuniformity as a result of the screen printing process. Many glazing/bonding temperatures and times up to 475 °C for 75 min. were explored in an attempt to reduce the melt-viscosity with no success. Furthermore, it was noted approximately 6 months after vacuum testing, that each of the 9 metalized, frit-sealed windows showed an additional failure mode due microcrack formation within the electrical feedthrough traces (Fig. 4.10). The exact cause is unknown, although it is presumed to be a result of a CTE mismatch. As an alternative, commercially pro-

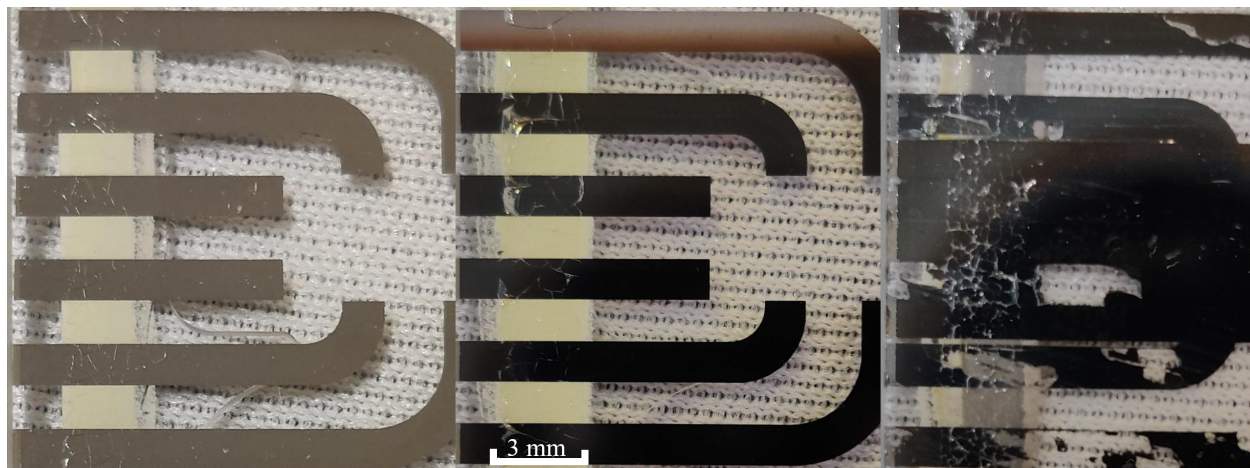


Figure 4.10: *Electrical feedthroughs exhibiting various levels of cracking under the frit bond.*

duced weldable, UHV multipin electric feedthroughs are available and relatively affordable for prototyping applications¹⁰¹.

4.2.2 Indium Seal

A glass-metal hermetic seal must be formed between the metal envelope and window assembly after thin film deposition. Thermo-compression seals between metals and glasses may be formed using indium or InAg alloys as an intermediary. A low-temperature bonding technique was planned following the glass-glass sealing processes outlined by Argonne National Lab and the LAPPD project for the production of MCP PMTs^{28;56}. Pure indium melts at 157 °C and readily wets to bare glass. Other constituents, such as silver, may be added to form an alloy, which alters the physical characteristics of the bond depending on the mass ratio. For example, an alloy of 3% Ag and 97% In has three times the tensile strength of pure In, with a liquidus temperature of 143 °C, but exhibits lower wettability¹⁰². It has been shown that In will bond to bare or metalized glass in minutes at temperatures as low as 20 °C if sufficient force is applied — 195 N/cm for 1.5 mm diameter indium wire. Bond strength does not change appreciably with higher processing temperatures and longer times for metalized samples under high compression load, although elevated temperature does improve strength under reduced compression (147.7 N/cm), likely due to increased malleability for physical bonds and diffusion rate for chemical bonds²⁸. Indium is prone to detrimental oxidation, forming a passivation layer 5-10 nm thick at room temperature, which reduces the diffusion rate or acts as a complete barrier. To improve bond strength, it is recommended that the oxide layer be removed by etching with 10% HCl for approximately 1 minute, rinsing in DI water and isopropyl alcohol to remove residual acid¹⁰³.

Alternative methods have been used to effectively remove oxides from the bonded area but require higher processing temperatures. To do this, a gap of 50 μm is maintained between the substrates. An In or InAg wire is placed around the sealing edge and heated well beyond the melting point as part of the vacuum bakeout process. Molten indium wicks into the gap via capillary action, and the oxide is left behind. This process occurs over a 24-34 hr period, after which the temperature is reduced for further processing, and a solid seal is formed³². Figure 4.11 shows an example of indium diffusion bonding of two NiCr/Cu coated borosilicate substrates²⁸.



Figure 4.11: *NiCr/Cu coated glass with a pure In diffusion bond formed at 350 °C over 24 hr²⁸. Over this period, indium diffused through the entire 400 nm film.*

A combination of elevated temperatures (80-130 °C) and light compression of 2 mm thick, pure In preform gaskets were planned for Catoptric device assembly which never came to fruition. Investigations of pure In bonds formed between both bare and NiCr/Au metalized borosilicate paired to stainless steel coupons at 120 °C revealed that indium weakly bonds to untreated stainless steel. Such bonds are likely mechanical in nature rather than chemical, and may not reliably seal a vacuum device without compression. To form a strong, reliable bond to stainless steel, InSn alloys are typically used alongside flux to remove surface oxidation¹⁰⁴, however fluxes are generally incompatible with UHV environments. In the future, it is recommended that stainless steel components receive the same NiCr/Cu metalization as the glass components to ensure hermeticity.

4.3 Dynode Fabrication

The total dynode assembly measures 8 mm wide, 7.5 mm tall, and 151 mm long with 150-300 μm features running the full length. Dynodes are typically made of a UHV-compatible sheet metal — either formed at the mm-cm scale or etched at the μm scale. Due to the high aspect-ratio geometry and vastly different feature sizes of the dynode structures, traditional machining techniques were not possible. Two non-traditional approaches were taken, but challenges in dynode fabrication for the reflective photocathode PMT persist.

4.3.1 Aluminum

The first dynode stages were fabricated from aluminum through wire electrical discharge machining (EDM) to create the small variable feature sizes required by the Catoptric design. Following fabrication, the bare aluminium dynode stages were type-3 hard anodized, as seen in Fig. 4.12, and then metalized by RDT, Inc.

In theory, the 150 μm oxide layer would form an electrically insulating barrier between the surface of the dynode stage and the conductive aluminum substrate after which conductive layers could be deposited to serve as electrodes. Initial continuity testing revealed that the stages were electrically insulated from each other. Following these tests, the substrates were loaded into an electron-beam (E-beam) evaporator and 50/50/100 nm thick Al/Ti/Au layers were deposited. After deposition, continuity testing revealed that the layers were no



Figure 4.12: *The metalized aluminum wire-EDM dynodes. The metalized surface provided low resistivity across the electrode but could not maintain isolation between electrodes.*

longer electrically isolated. It is hypothesized that the porous oxide layer (Fig. 4.13) was not sufficient for such small features once metalized. Additionally, the porous aluminum oxide would likely trap a substantial amount of contaminants which may outgas and poison the PMT vacuum and sensitive materials. Alternative methods of surface oxidation may be studied in the future, should it be warranted. Methods of sealing anodized aluminum are common, but may not be compatible with the passively-pumped vacuum environment of a

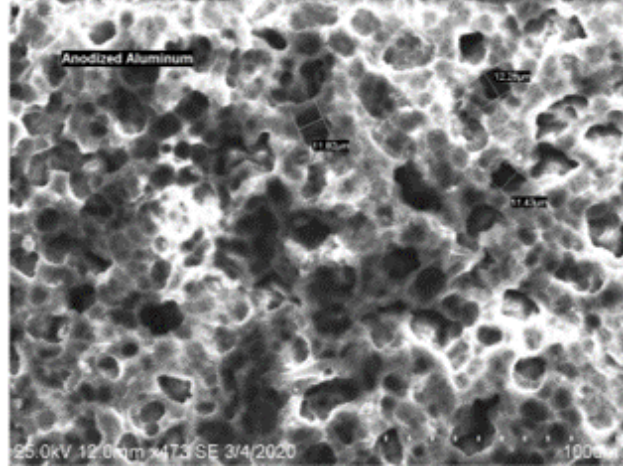


Figure 4.13: *The porous oxide formed after anodization. Image courtesy Taylor Ochs, RDT.*

PMT. Atomic layer deposition (ALD) of dense Al_2O_3 films is also common. ALD technology enables the deposition of highly conformal films over relatively large areas. Recently, 185 nm ALD Al_2O_3 films with a post-anneal dielectric breakdown strength of 6.1 MV/cm have been reported¹⁰⁵. Whether these films are mechanically robust enough for this specific application is unknown, and further research is left for future work.

4.3.2 3D Printed PerFORM

A second approach to dynode fabrication used stereolithography (SLA) additive manufacturing (AM), also known as SLA 3D printing. A resin impregnated with ceramic particles, Somos PerFORM, was chosen for its tight-tolerance capability, electrical properties, purported low outgassing (personal communication, April 1, 2020), and high temperature resistance¹⁰⁶. An initial 50 nm/200 nm Ti/Ni deposition carried out by RDT, Inc. on the PerFORM substrates resulted in high resistances, ranging from 200-1000 Ω across the dynode surfaces. An additional low-phosphorus ENIG electroplating process reduced resistance to approximately 15 Ω along the dynode length (Fig. 4.14). The electroplated electrodes were prone to flaking, likely caused by both poor surface adhesion and stress induced when deforming the material. This was partially rectified with additional process characterization and control. The PerFORM material exhibited poor temperature resistance during testing. Some dynode

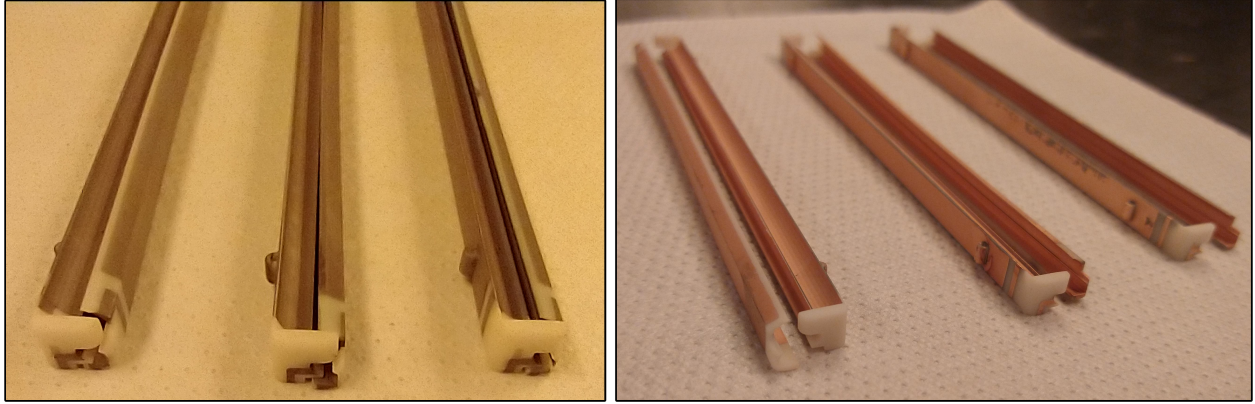


Figure 4.14: *The metalized PerFORM dynodes Left) after e-beam evaporation and Right) after the ENIG process.*

samples warped under their own weight at temperatures as low as 90 °C. It was therefore concluded that the material was not suitable for the dynodes, as it could not be properly degassed at higher temperatures. It is unlikely any polymer-based material is suitable for static UHV or HV environments with sensitive materials such as alkali-antimonides. Typical UHV-compatible polymers, especially AM materials, such as PEEK, PTFE, and Kapton are quite permeable, and are often a continuous source of free-radicals under UHV conditions⁵⁸. Many of these materials can and should be baked from 150-400 °C prior to use in UHV, but the outgassing rates of water and other atmospheric species are inevitably more substantial than those of metals and ceramics⁵⁸.

4.4 Conclusions and Future Work

Fabrication of the metal housing is relatively straightforward and envelopes may be produced through typical machining processes, however milling and polishing billet stainless is quite expensive. Future iterations for large production quantities should be formed from pre-polished stainless steel sheet. Modern robotic forming techniques have made formed parts accessible in prototype quantities, circumventing the engineering and fabrication investment otherwise needed to produce press forms.

Glass-glass bonding of borosilicate substrates using screen-printed frit paste was inves-

tigated. The formation of robust, hermetic glass seals was difficult to achieve, especially over large boundaries due to minor thickness nonuniformity. The 1.1 mm frame substrate was fragile and prone to breaking during vacuum testing. Metal traces cracked over time under the bonded frit in moderate indoor environments, likely due to a CTE incompatibility. Commercially produced UHV electrical feedthroughs are available, however, they may not be congruent with the current PMT design. Alternatively, other frit compounds from manufacturers such as Schott may be explored.

Preliminary investigations of glass-metal indium bonds show that seals are reliably made over large boundaries using a combination of heat and compression. Hermetic bonds may be created on both bare and metalized glass surfaces, though the latter offers additional reliability and strength with the use of silver alloys. Further exploration of indium bonding to stainless steel is needed, however NiCr/Cu metalization of stainless steel surfaces should offer similar performance to that of metalized glass substrates.

The current Catoptric dynode design is complex from both a fabrication and processing standpoint. Deposition and handling in UHV, while maintaining discontinuity between electrodes is expected to be a substantial challenge if functional dynode structures can be produced. Still, the design may be viable if electrodes can be fabricated using a metallic wire-EDM substrate. Unexplored anodization-sealing and ALD processes may yet produce a sufficiently resistive surface film. Alternatively, a similarly sized wire-mesh or metal channel plate style dynode assembly may offer improved collection efficiency and gain performance while enabling simpler UHV processing given sufficient development. A mock Catoptric PMT assembly may be seen in Fig. [4.15](#).

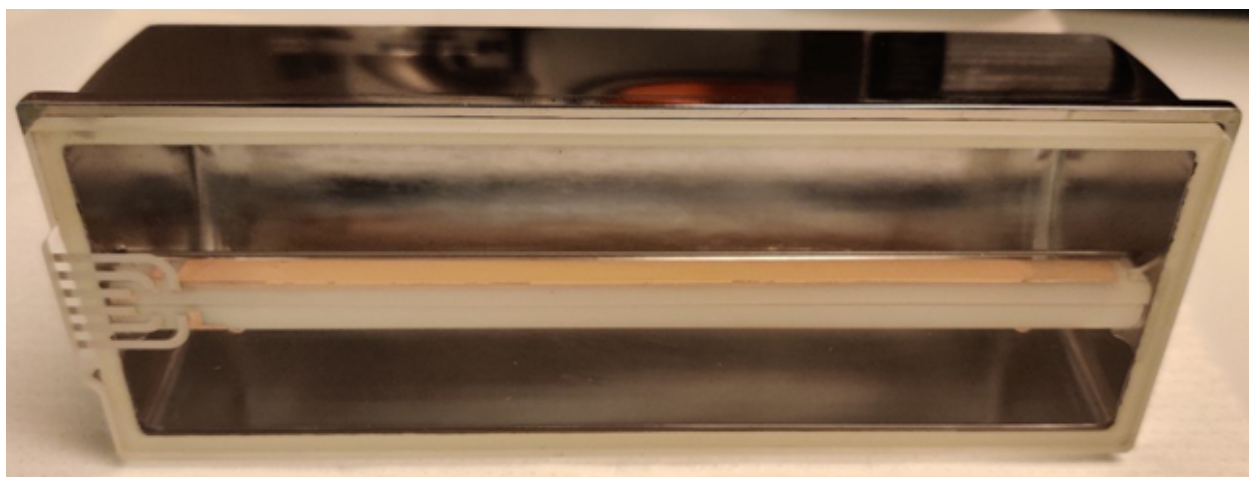


Figure 4.15: *Fabricated components are shown in a mock Catoptric PMT assembly.*

Chapter 5

Vacuum Chamber Design

5.1 Vacuum Technology

A large (approximately 56 l) 304 stainless steel vacuum vessel was designed and constructed to enable UHV thermal deposition and PMT assembly. The chamber utilizes multiple vacuum pump and analysis systems, as well as custom translators, manipulators, and bellows to facilitate substrate translation and assembly. Resistive and lamp-based heaters provide thermal control for the sources and substrates, while type K thermocouples are used to monitor the local temperatures. The design and functionality of these subsystems are described herein.

5.1.1 Vacuum Pumps

Four vacuum pumping technologies were used to achieve a UHV environment within the chamber. Namely, two dry scroll pumps, with a total pumping speed of 4.4 l/s, two high-compression 250 l/s turbomolecular pumps, two titanium sublimation pumps (TSPs), and a 410 l/s ion pump were used. The chamber flanges utilized oxygen-free, copper ConFlat gaskets to maintain a low leak-rate seal. Manual UHV gate valves were used to separate the external atmosphere from the vacuum chamber, isolate turbopumps from the chamber when needed, and to isolate the ion pump during alkali metal deposition.

Roughing Pumps Dry scroll pumps, which are specifically designed for UHV or chambers requiring low hydrocarbon contamination, were chosen to bring the chamber to rough vacuum. Inside the scroll pump are two Archimedean spirals, one of which is nested within the other. The nested spiral translates in a cyclical motion within a fixed outer scroll, while PTFE seals trap the air which is compressed as it moves towards the center. There, the compressed gas is expelled to atmosphere as illustrated in Fig. 5.1. The pump is “dry”

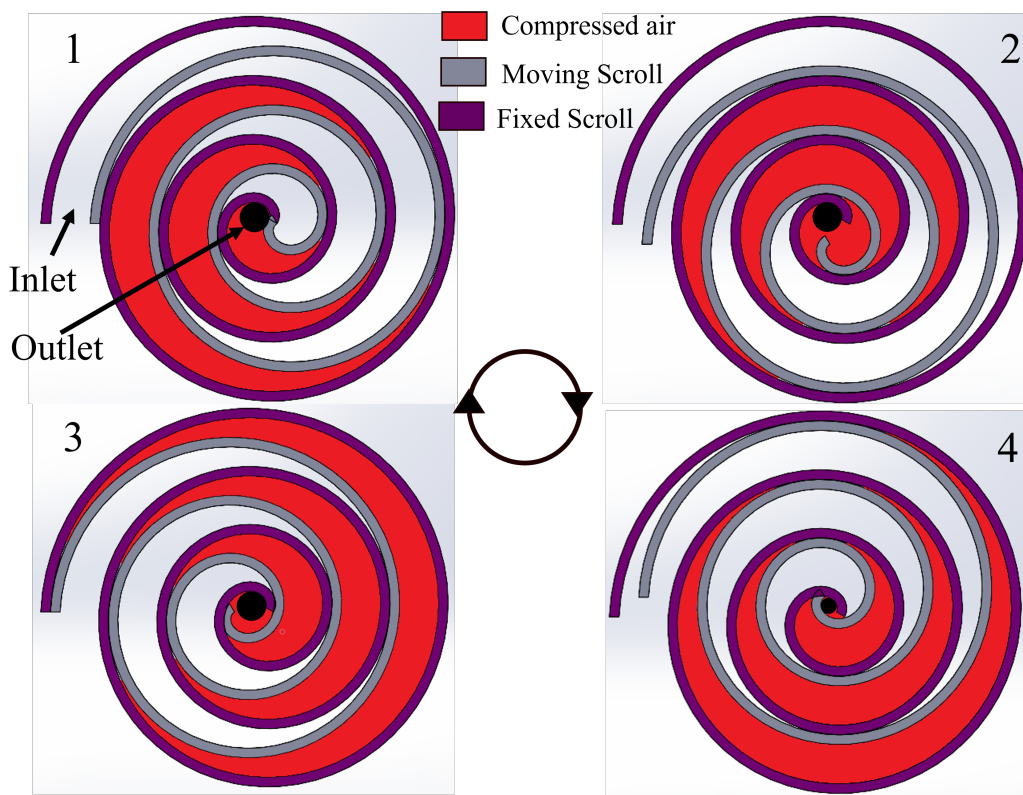


Figure 5.1: *The operational cycle of a scroll pump.*

because the bearings are isolated from the pumping volume, and grease is not used to seal the cavity. This is imperative for UHV systems which cannot be contaminated by grease or oil vapor produced within typical roughing pumps that may backstream to the primary pump. Roughing pumps can achieve chamber pressures approaching 10^{-3} torr, and are used in conjunction with turbomolecular pumps to achieve higher vacuum.

Turbomolecular Pumps Turbomolecular pumps, or turbo pumps, are the most commonly used primary pumps for HV and UHV systems. To achieve UHV within the fabri-

cation chamber, high-compression ratio TwisTorr 304FS turbo pumps were chosen¹⁰⁷. The rotating assembly within a turbo pump consists of multiple axially-mounted turbine stages (Fig. 5.2). The stages rotate in unison up to 60,600 rpm with the aluminum blades suspended

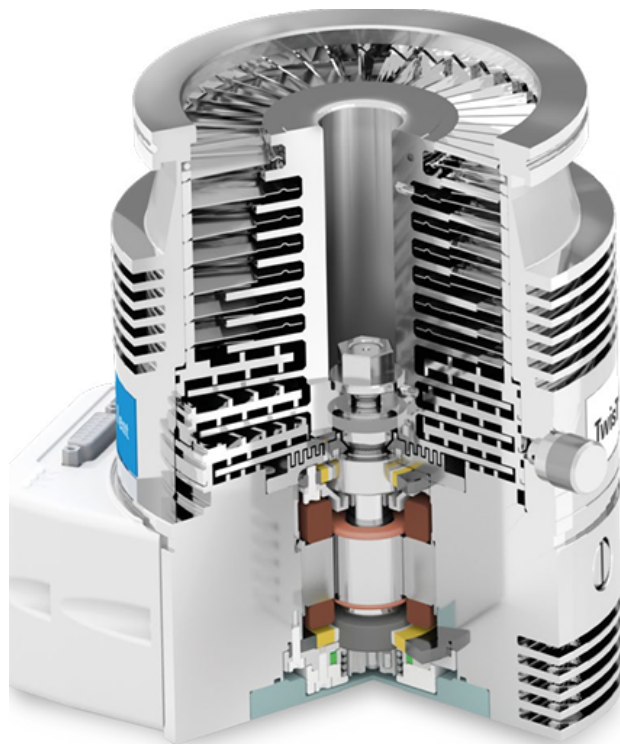


Figure 5.2: A cutaway of an Agilent Technologies turbomolecular pump similar to those installed on the UHV deposition and assembly chamber. Image courtesy Agilent Technologies, Inc.²⁹

by magnetic and/or ceramic bearings. Under vacuum, atoms and molecules populating the chamber interior are in the molecular flow regime and no longer act as a fluid¹⁰⁸. However, through random surface and particle interactions they may reach the pump inlet. Those that reach the first stage of the pump may be struck by one of the rotating blades and given additional momentum in the axial direction of the turbine. Those that achieve the appropriate trajectory are accelerated by subsequent stages, eventually being ejected from the system to atmosphere by the roughing pump. High-compression turbomolecular pumps, such as those used on this chamber, have an ultimate pressure below 10^{-10} torr¹⁰⁷. At pressures lower than this, the turbo pump(s) should be valved off from the chamber to prevent a backflow of molecules into the chamber. Although this level of vacuum is sufficient for bialkali thin

film growth, additional pumps were used to increase the pump rate and lower the partial pressures of reactive contaminants.

Titanium Sublimation Pumps Titanium sublimation pumps use high current to evaporate titanium from a filament onto chamber surfaces. Highly reactive titanium acts as a getter to absorb and trap vacuum contaminants. TSPs are highly advantageous in the lower UHV regime where H_2 is the dominant background gas. Two TSPs with theoretical hydrogen pumping speed of 1600 l/s each were added to the chamber. In this system, the two units, each composed of three individually controlled titanium filaments and a large-area shroud, may be operated separately. Power supplies operating at up to 800 W at 55 Amps intermittently heat filaments coated in titanium which evaporate and coat the large-area fins (Fig. 5.3)³⁰. The titanium then cools and serves as an effective getter, adsorbing reactive molecules present within the vacuum atmosphere.

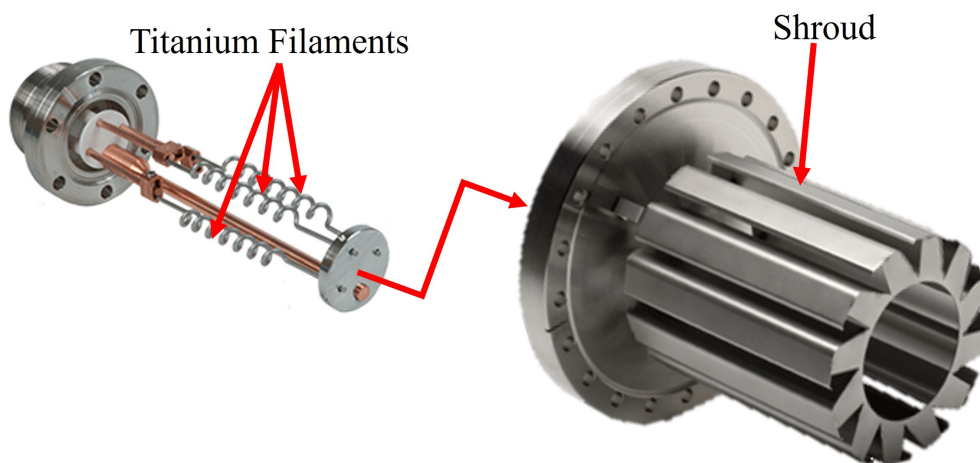


Figure 5.3: *The TSP and shroud assembly used to maintain UHV. Images courtesy Gamma Vacuum³⁰.*

Ion Pump An Agilent VacIon 500 StarCell combination ion pump was included to further increase the pumping rate of the chamber. Additionally, the operational mechanism of the ion pump allows for accurate pressure measurement. The selected model incorporates one of

the aforementioned TSPs within its housing. Ion pumps use 3-7 kV potential and a 0.1-0.2 T magnetic field to form a plasma of ions and free electrons. The ions accelerate towards the Ti cathode and may be pumped in one of two ways:

1. Smaller ions (e.g. H_2) may penetrate the cathode surface and diffuse into the Ti where it is trapped.
2. Titanium is sputtered onto the anode walls by larger ions striking the cathode. Reactive ions such as oxygen and nitrogen impinging the cathode surface are permanently gettered at the anode or other internal pump surfaces.

A cutaway of a typical ion pump design featuring the primary components may be seen in Fig. 5.4. The StarCell cathodes feature a unique geometry (not shown in Fig. 5.4) which

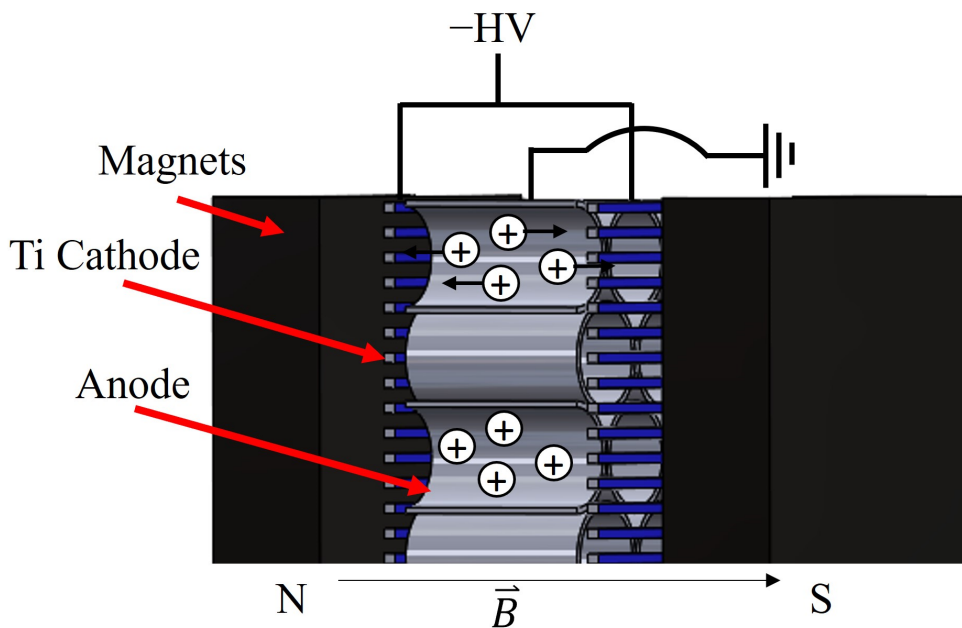


Figure 5.4: *The internal components of an ion pump. Ions formed in the plasma are accelerated by the electric field and are pumped by Ti gettering.*

increases the amount of Ti available for sputtering while improving inert gas stability. Some inert gasses may be temporarily removed by the ion pump through a “wall-papering” effect, wherein sputtered Ti is deposited over top. However, upon heating the ion pump, these nonreactive gasses may diffuse to the surface. Therefore, it is still important to use a turbo

pump to remove inert residual gasses^{58;109}. Because of the high voltage required to form the plasma, it is important that conductive, diffusive contaminants such as the high vapor pressure alkali materials do not have a direct line into the ion pump. To mitigate this issue, the ion pump was placed out of the line-of-sight of the sources. This does, in effect, reduce the molecular conductance and consequently the pumping speed.

All units, besides the roughing pumps and one TSP, are controlled and monitored from the computer over RS232 using Agilent Technologies' A-Plus software¹¹⁰. It is recommended that another method of computer-based control is implemented in the future, as A-Plus limits monitoring, control, and data recording to a single device at any given time.

5.1.2 Vacuum Analysis Equipment

Dual-Range Vacuum Gauge Prior to and during deposition, it is imperative to monitor atmospheric conditions within the chamber. To do this, three devices are used. To monitor vacuum levels from atmospheric pressure to 3.8×10^{-9} torr, an Agilent FRG-702 Pirani/inverted magnetron combination gauge was used¹¹¹. From atmospheric pressure to HV, the Pirani gauge uses a heated wire to measure the pressure-dependent thermal conductivity. In the HV/upper-UHV regime, the gauge transitions from Pirani gauge operation to the inverted magnetron gauge. Similarly to the ion pump, a plasma is formed within the gauge and the current flow from the anode to the cathode is measured to measure the pressure. It is understood that the inverted magnetron device is a cold cathode gauge (CCG) which does not operate as effectively if contaminated by a deposition source such as K or Cs. For this reason, it is generally preferable to have a Bayard-Alpert, hot cathode gauge (HCG) device, which due to its temperature is not readily contaminated during deposition. Due to stocking issues at the time of delivery, this was not feasible.

Residual Gas Analyzer Beyond the range of the combination gauge, both the residual gas analyzer (RGA) and ion pump can be used. The RGA100 from Stanford Research Systems operates over a pressure range of 10^{-4} torr and beyond¹¹². If calibrated correctly, the device can accurately read total and partial pressures of molecules ranging from 1 AMU

to 100 AMU from 10^{-4} - 10^{-11} torr. Two thoria-coated iridium wires are heated to produce free electrons which are accelerated towards a mesh anode. Some strike the anode and are absorbed; others pass through the mesh and oscillate back-and-forth, increasing the probability of an ionization. Ions formed with the concentric anode are focused to the mass-filter entrance. The device uses a cylindrical-rod type quadrupole for mass filtering and a Faraday cup detector to directly measure the ion current. An electron multiplier is available on some to increase partial pressure sensitivity to 10^{-14} torr, if needed¹¹². Software provided by SRS is used for mass calibration, averaging, and live partial pressure readout as well as data logging throughout deposition and assembly. Mass scan-rate and averaging may also be controlled, and when used correctly, provide accurate partial pressure readings in the UHV range.

5.2 PMT Vacuum Fabrication Equipment

5.2.1 Antechamber

To prevent primary chamber exposure to atmospheric contaminants during repeated deposition/assembly cycles, an antechamber, or load-lock, was designed. The load-lock is simply a four-flanged chamber. The transverse flanges are reserved for vacuum equipment, while the remaining axial ports are used for substrate transfer. The outer flange has a viton-sealed door with a viewing window. Individual components were to be loaded into the antechamber, the door shut, and the chamber pumped to between 10^{-8} torr and 10^{-9} torr. Once at an appropriate level, the valve to the main chamber would be opened. Initially all components were designed to fit within the antechamber simultaneously, but other design constraints ultimately removed this functionality. To overcome this, 2 antechamber pump-downs would be required which adds time to the overall process. Ultimately, the antechamber was never incorporated, as repeated component loading without additional internal maintenance was never required. Figure 5.5 shows the proposed component layout for the load-lock, which was fabricated, but never utilized.

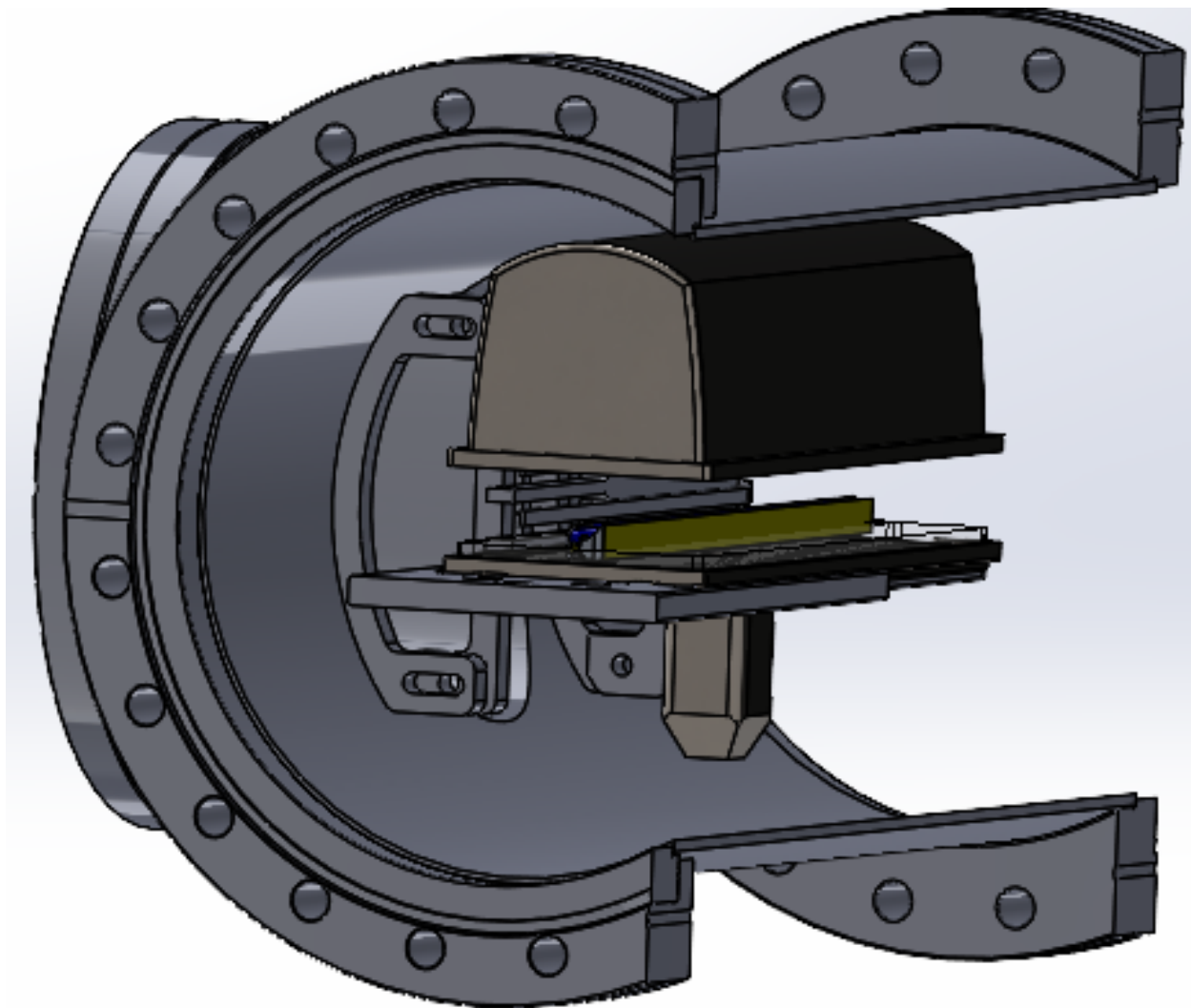


Figure 5.5: *A cross-sectional view of the antechamber containing the PMT components.*

5.2.2 Sample Manipulation

Axial Translator To maneuver the substrates throughout the chamber, multiple UHV manipulators were included. For axial translation, a magnetically-coupled translator from UHV Transfer Systems, Inc. was installed on the far end of the system. The translator is coupled both linearly and rotationally. A custom end-effector was designed to transport the cathode envelope, window carrier, and dynode carrier. By rotating the magnetic housing, an internal shaft is turned and allows for 1 in. vertical sample translation — enabling substrate transfer between the three locations within the load-lock and primary chamber. Figure 5.6 shows the axial translator end-effector prior to polishing.



Figure 5.6: *The end-effector mounted the axial manipulator prior to polishing. The fork design enables stable transfer of the window, dynodes, and PMT envelope.*

Transverse Manipulator Another linear translator from Mcallister Technical Services is mounted transversely to the chamber. This translator carries the deposition stage and controls substrate positioning above the evaporation sources. Rather than magnetic coupling, the device uses edge-welded bellows paired to linear bearings and is moved using a lead screw.

Wobble Stick A third device from UHV Design, known as a wobble stick, is also mounted transversely near the assembly area and may be used for dynode and photocathode substrate manipulation. The wobble stick maintains a wide range of motion using both welded bellows and magnetic coupling. The wobble stick is capable of a 22° tilt angle, 360° rotation, and has a spring-loaded “trigger” which manipulates a custom set of tweezers used to grab and align the dynodes into position. Figure 5.7 shows a cross-sectional model of the assembled device.

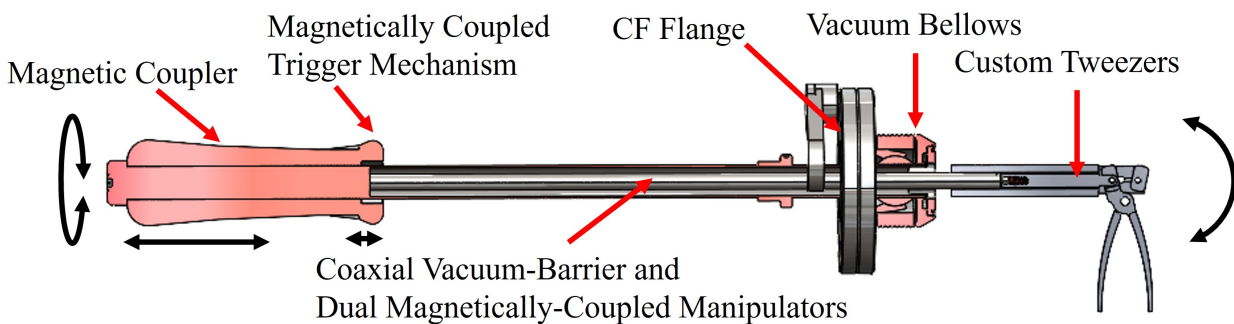


Figure 5.7: *A cross section of the wobble-stick/tweezer assembly illustrates how the assembly functions in UHV.*

5.2.3 Thermal Deposition Equipment

Linear Effusion Cells Thermal physical vapor deposition of the photocathode and secondary emissive materials is carried out using non-epitaxial effusion cells. These sources were chosen for their higher capacity and repeatability over simpler boat sources or specialty chromate-based sources, such as those from SAES. A custom linear effusion cell array was designed by SVT Associates, Inc. based on the source materials, substrate geometry, and uniformity requirements outlined by Kansas State University Researchers. The three units each consist of two primary chambers with individual thermal control capabilities. The lower section is a crucible surrounded by filament heaters and thermal insulation, which contains the bulk material. The upper section is a manifold encased by filament heaters and radiative thermal insulation that linearly distributes the deposition material.

The deposition material held in the crucible is heated to the point of vaporization under UHV. The deposition rate is controlled by the temperature of both the crucible and manifold and is further limited by an orifice. By increasing crucible temperature, the internal pressure increases. Deposition material exits through the orifice and enters the distribution manifold. The manifold, held at a temperature slightly above that of the crucible to avoid material condensation, has precision-machined holes which emit material with a flux distribution tuned for the cathode geometry, as seen in Fig. 5.9. Two fill ports (not shown in Fig. 5.10), accessed outside the chamber, allow for the transfer of alkali material into individual crucibles.

Deposition Stage A custom deposition stage was designed and fabricated to facilitate deposition process control. The stage is mounted to a transverse linear-manipulator to position the substrates over deposition sources. The deposition stage may simultaneously hold the photocathode and dynode substrates, with the latter mounted to a carrier. Two AC-powered, quartz lamps controlled using variable transformers are used for dual-zone thermal management. Each zone uses type-K thermocouples to monitor approximate substrate temperature. Antimony deposition on stainless steel was measured internally by a QCM from McVac Manufacturing. A tooling factor calibration deposition was performed and quantified

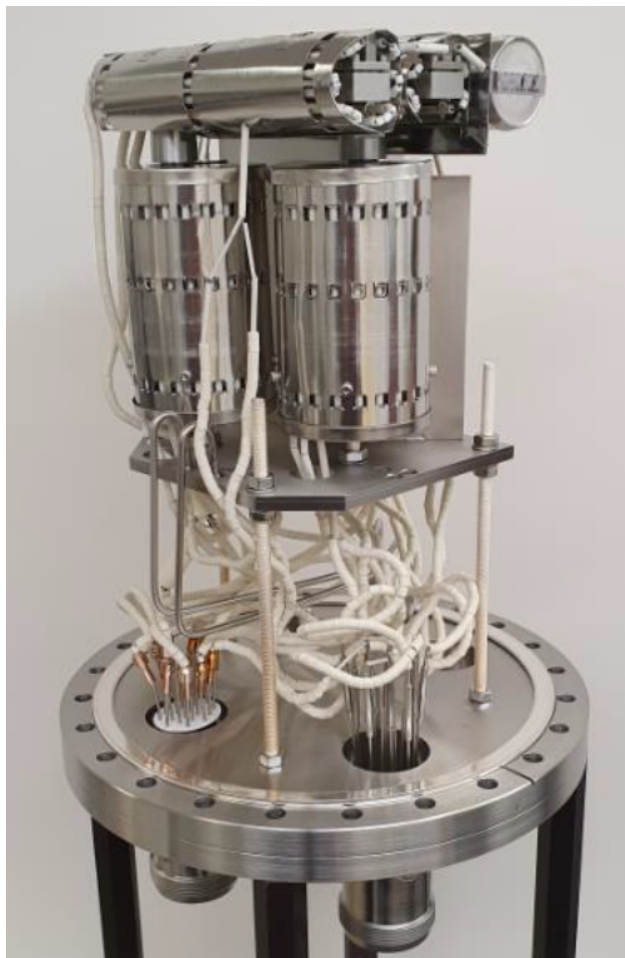


Figure 5.8: *The effusion cell assembly with additional radiative insulation and a cooling plate to reduce thermal cross-talk.*

using a Dektak 150 surface profilometer for accurate measurements during cathode deposition. Optimal thicknesses for both alkali materials are determined by monitoring the peak photocurrent emitted by the thin film.

Photocurrent Monitoring Equipment To measure photocurrent, both fixed and variable wavelength light sources are used in conjunction with a Keithley 6487 combination picoammeter/voltage source. The light sources are coupled to deposition-stage mounted collimators through two UHV compatible, 1000 μm UV/VIS multimode fiber optic cables within the chamber. Each vacuum fiber optic is then coupled to an SMA fiber-optic feedthrough near the deposition-source base. A 1000 μm UV/VIS fiber optic cable is routed from the

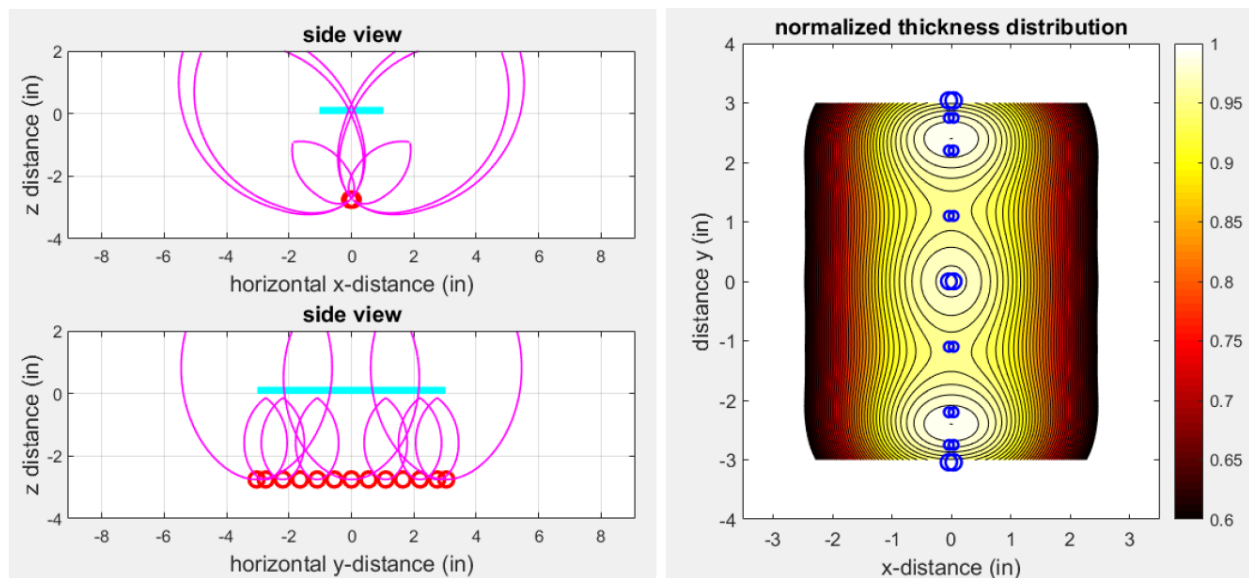


Figure 5.9: *The simulated source distribution from the effusion cell manifolds. Each manifold shares the same emission-hole pattern. Simulation courtesy SVT Associates.*

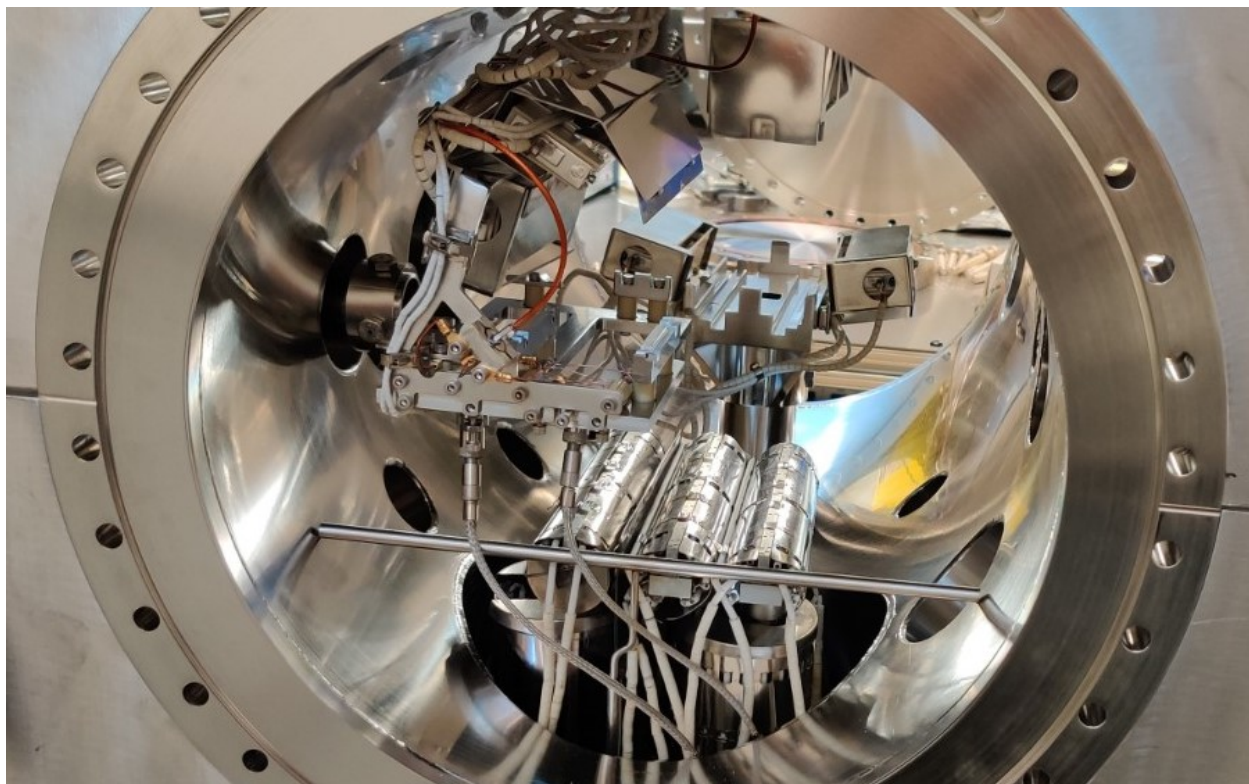


Figure 5.10: *The deposition stage provides dual-zone heat and photocurrent measurement for photocathode/secondary emitter deposition.*

variable-wavelength source and connects to the feedthrough. The fixed-wavelength source may be directly connected to the feedthrough to limit coupling losses. The UHV-collimation assembly provides a spot size of approximately 5 mm at the photocathode. Figure 5.11 shows the optical-source internal calibration process.

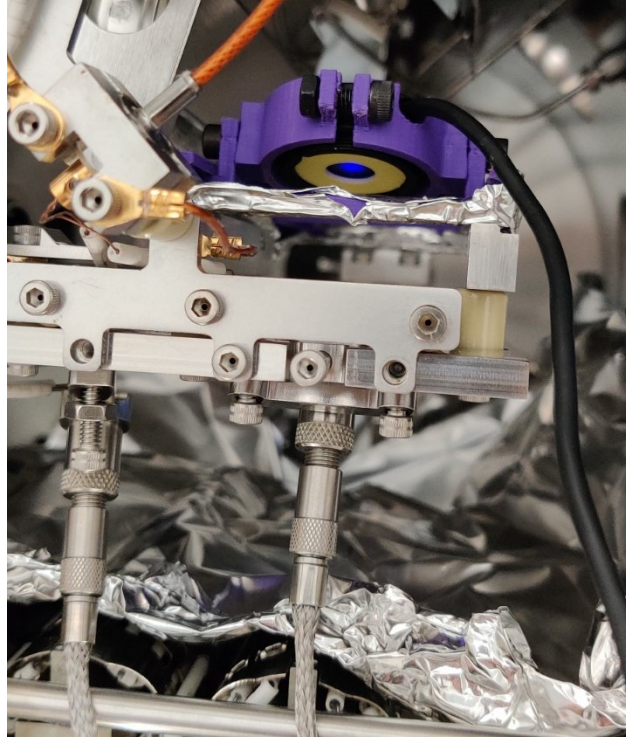


Figure 5.11: *An S120VC UV-extended photodiode from Thorlabs is placed in the deposition location to measure the optical power transmitted through the fiber-coupling.*

The light sources are calibrated prior to measurement using a Thorlabs S120VC photodiode and PM100USB power meter. The fixed-frequency source is a BOLB 275 nm, 100 mW radiant flux, 35° FOV LED¹¹³. The LED is mated to a heat sink to ensure stability and extended lifetime. Modular SM1 connectors from Thorlabs are used for optical coupling to filters, fiber optic, etc., and may be seen in Fig. 5.12. Optical power for the 275 nm LED assembly was quantified internally and externally using a variety of neutral density (ND) filters and drive currents to calibrate the transmission factor.

The variable-wavelength source pairs a 100 W APEX2-XE xenon arc lamp to an Oriel Cornerstone™ 260 1/4m UV-VIS monochromator, as shown in Fig. 5.13. The broadband light source is first passed through the filter wheel, then to one of two diffraction gratings

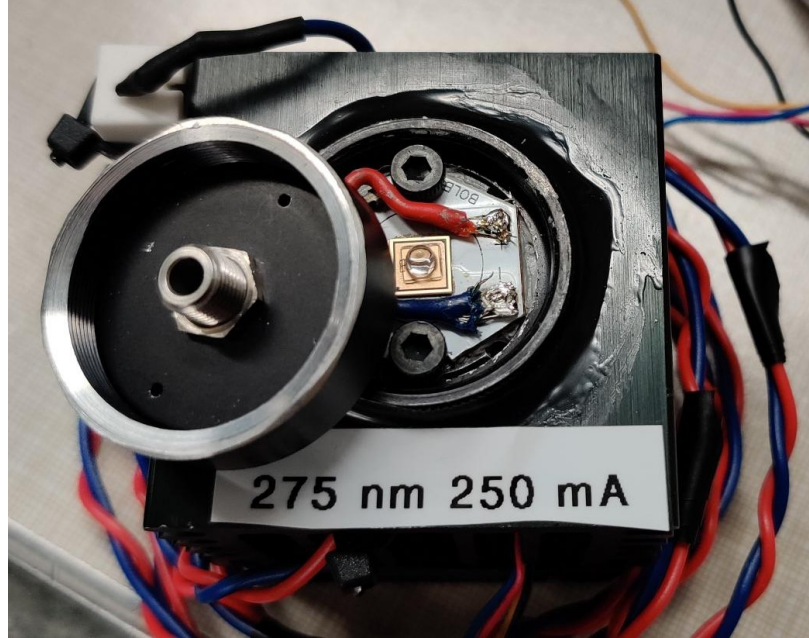


Figure 5.12: *The 275 nm LED light source alongside an SM1 adapter for fiber optic.*

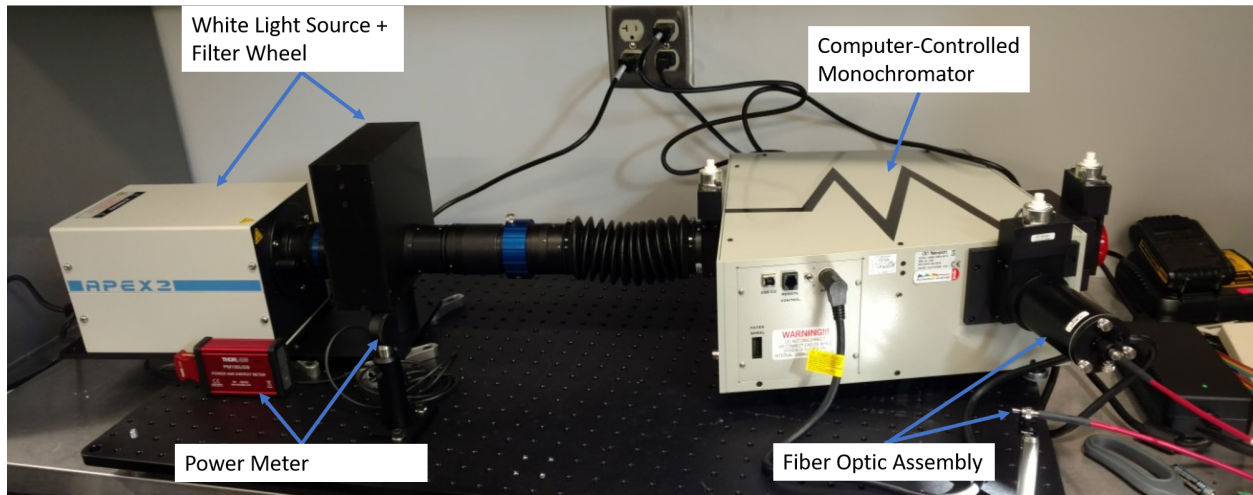


Figure 5.13: *The broad-spectrum light source and monochromator provide a variable-wavelength light source for photocathode characterization.*

within the monochromator, and finally directed to one of two output ports. Grating 2 has the highest efficiency throughout the pertinent spectral range, and was thus chosen as the sole grating used during measurements. The variable slit-width, which defines the bandpass, is controlled through USB using a standalone application or by interfacing the application through Python. A fixed bandpass of 20 nm was chosen to maximize the radiant flux at

the substrate. Care must be taken to omit higher orders of radiation when performing spectral analyses with diffraction gratings. A 10-GCA435 longpass filter with a 435 nm cut-on wavelength from Newport was chosen to filter out higher-order radiation while performing measurements beyond 450 nm.

Using the S120VC calibrated photodiode, internal and external power as well as the transmission factor were measured in 5 nm intervals over the entire spectral range. Prior to each deposition, an external power measurement was taken to ensure source stability and to quantify the optical power at the cathode. These values were then multiplied by the transmission factor to calculate the internally transmitted optical power. This serves as a best-approximation, as there is no way to ensure the internal transmitted power is not affected during deposition. Figure 5.14 illustrates the standard internal output and transmission factor for the variable-wavelength source.

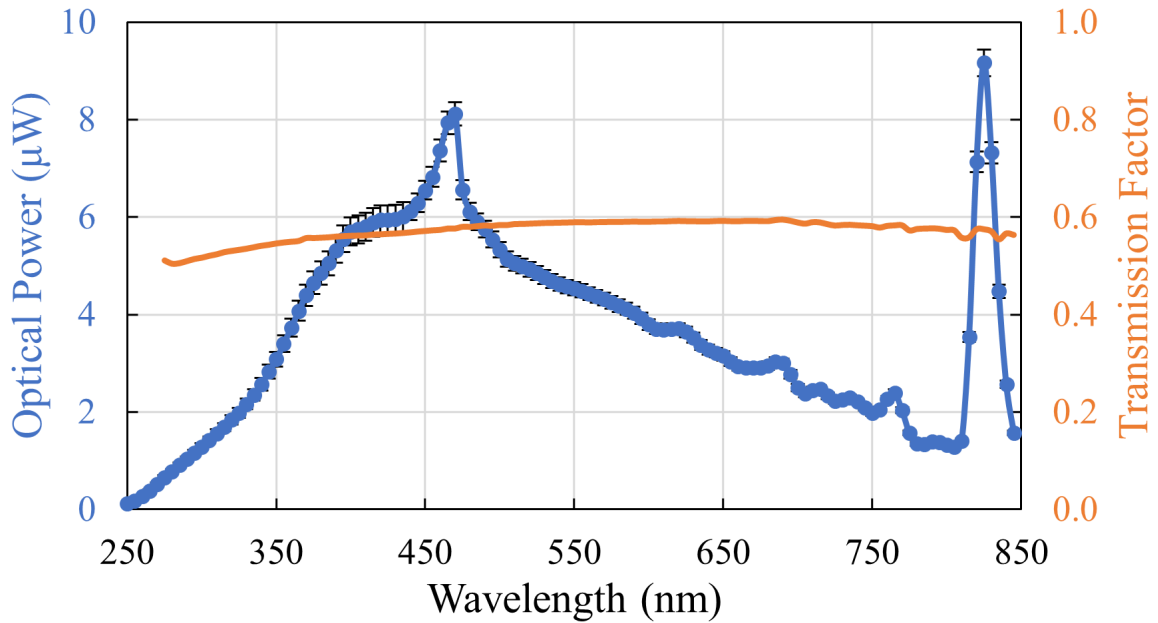


Figure 5.14: *Internal optical power and transmission factor for a spectral measurement of the variable-wavelength light source.*

For current measurement, a twisted pair inside a knitted wire mesh sleeve extends from the picoammeter voltage supply. The sleeve is connected to the outermost shield of the triaxial cable which serves as a chassis ground. A wire routed from the LO connection has

been soldered to the “input low”, middle sheath of a triaxial cable. The voltage supply HI connection is passed into the chamber using the internal conductor of a BNC feedthrough and polyimide insulated BNC cable. Finally, the “input high”, innermost conductor of the triaxial cable is connected to the outer conductor of the isolated BNC feedthrough and cable. A connection diagram can be seen in Fig. 5.15.

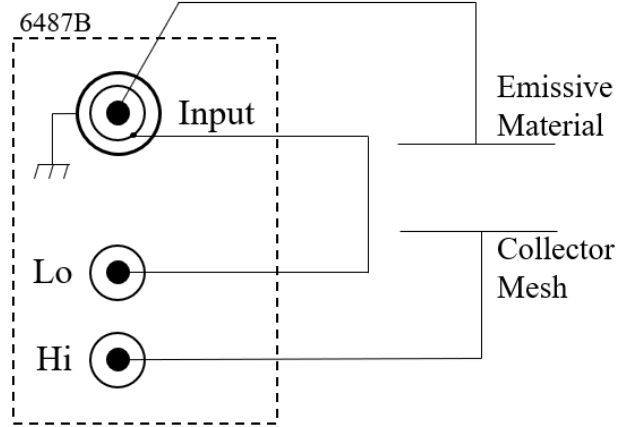


Figure 5.15: A diagram of the connections used for photocurrent measurement during deposition.

Inside the chamber, each substrate is electrically isolated from the deposition stage. Ceramic insulators hold metal substrate mounts, such that the substrates are electrically connected to input high on the picoammeter. Wire-grid collection anodes are formed between long screws in both substrate deposition locations and are held at a higher potential to attract emitted photoelectrons. Photocurrent emitted from the photocathode is measured by the picoammeter, and an integration window is used to effectively filter out noise induced by the AC-powered heat lamps and other external sources. Live photocurrent data is plotted and saved for real-time monitoring and post-processing. Using the wavelength-dependent equation below, QE may be calculated as

$$\eta_{QE}(\%) = \frac{hc}{\lambda e} \frac{I_k}{L_p} 100\% = \frac{1240}{\lambda} \frac{I_k}{L_p} 100\%, \quad (5.1)$$

where η_{QE} is the QE, h is Planck’s constant, c is the speed of light in a vacuum, e is the

charge of an electron, I_k is the photocurrent in units of A, and L_p is the incident radiant flux in units of W¹⁶. Both single-wavelength and wavelength sweeps may be taken *in situ* to monitor photoemission characteristics.

A multi-step calibration procedure ensures accurate measurement of low currents by the picoammeter. Initial measurements tested a 1 G Ω resistor in a project box. A TRX-BNC feedthrough was used, similar to the vacuum chamber setup. 5 VDC was applied from the picoammeter and significant noise was discovered in the signal with no averaging, no filtering, and the fastest integration setting regardless of shielding. One component of the noise was at 60 Hz frequency, but an unknown multiplicative component had a 1.5 Hz frequency regardless of the test configuration. The built-in Medium rank filter, 100 count moving average, and medium integration time were sufficient to provide accurate, stable readings. Similar results were found within the chamber with pumping and heating equipment active at each deposition location. Any noise was successfully filtered by the picoammeter, providing stable readings into the sub-nA range.

To test the photocurrent measurement process within the chamber, a small sample of 99.9% bulk silver was excited by the 275 nm LED. The sample was prepared by sanding the surface with 400 grit sandpaper to remove any oxide layer. The surface was then cleaned using an ultrasonic cleaner and 10% Citranox acid-based cleaner combined with distilled water. An acetone rinse followed by isopropyl alcohol ensured that no residual contaminants remained on the surface. The sample was immediately loaded into the vacuum chamber which was promptly evacuated to 3×10^{-7} torr. The sample was heated to approximately 230 °C. Operating the LED while heating, photocurrent rose slowly over the course of 30 minutes before plateauing. It is believed that some contaminants and a thin oxide layer were removed during this process, providing the surface conditions necessary for photoemission. The sample was allowed to cool, and a series of photocurrent measurements were taken to confirm silver's response to 275 nm light. First, the potential applied to the anode mesh was swept from 0 V to 300 V in 25 V increments with 655.4 μ W radiant flux impinging the silver sample to find the required collection voltage. Photocurrent began to plateau at 150 V, therefore further testing was performed with the mesh between 150-250 V. The results of

the voltage sweep are shown in Fig. 5.16.

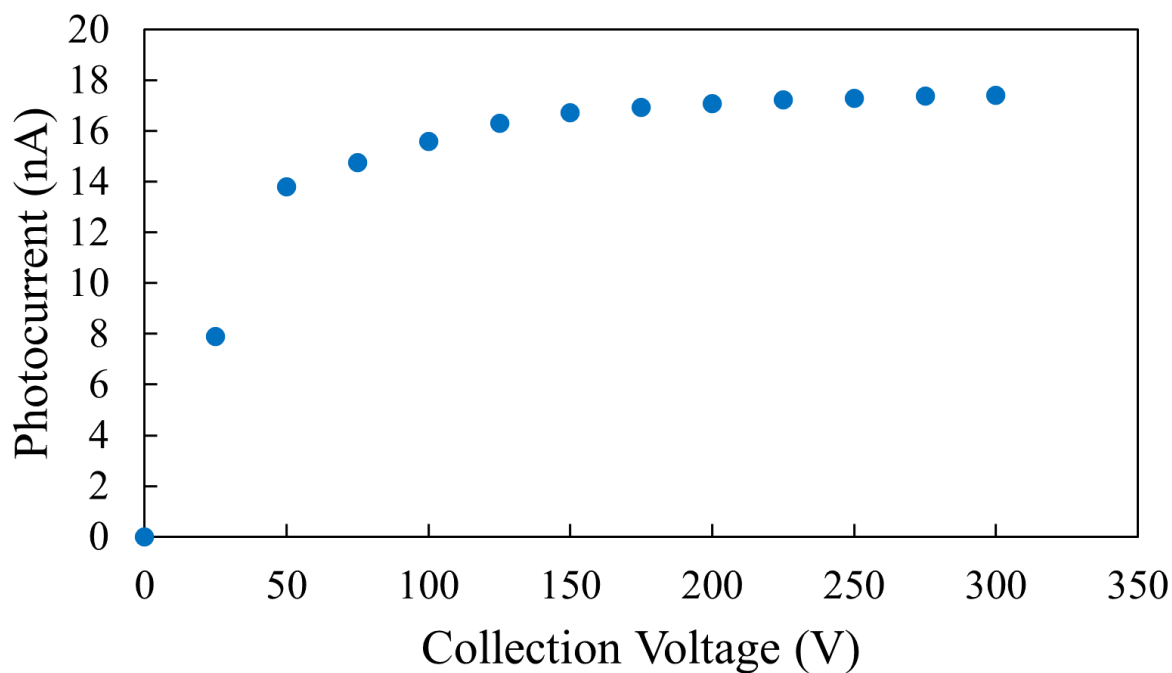


Figure 5.16: *A voltage sweep was performed to ensure all charge would be collected during photocurrent measurement. A collector-potential of 150 V was required.*

Varying radiant flux by reducing the current through the LED produced a very flat QE of approximately 0.0115% between 6.9 nA and 19.2 nA photocurrent. The results of the silver QE measurement may be seen in Fig. 5.17.

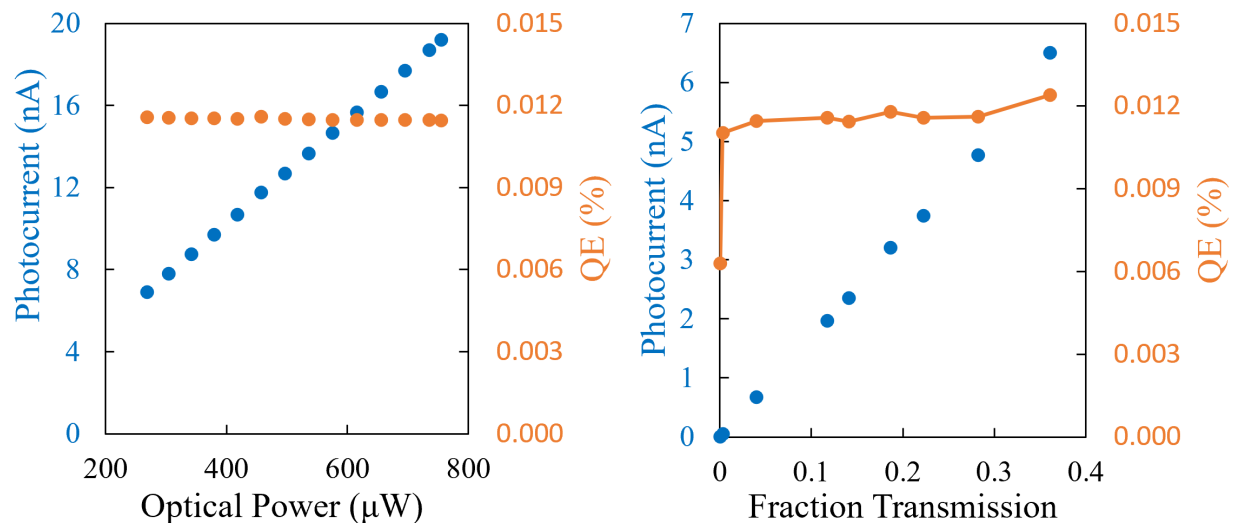


Figure 5.17: *Left) The photocurrent and QE as a function of the optical power modulated by changing the drive current. Previous testing showed that drive current is linearly proportional to optical power. Right) The photocurrent and QE are plotted against the fraction transmission as measured with various optical-density ND filters.*

5.2.4 Assembly Equipment

Final assembly of the device may be performed using a custom assembly stage composed of internal alignment tools, a manipulator, and heaters. PMT components are moved to the assembly station after deposition.

Internal Assembly Stage Two interior components machined from billet 316 stainless steel provide alignment for the PMT assembly. Two AC-powered lamp heaters allow for component outgassing, getter activation, and indium sealing. Type K thermocouples mounted to either side of the lower structure monitor temperature near the indium seal interface. Self-aligning features on the PMT components are further aided by the alignment structures designed into the assembly structure. The interior assembly structure was designed in such a way to mitigate virtual leaks. After the initial machining process, parts were first mechanically polished and then electropolished to limit the outgas rate within the chamber so as to improve the base chamber pressure. Figure 5.18 shows the assembly stage stack with PMT and manipulation components incorporated.

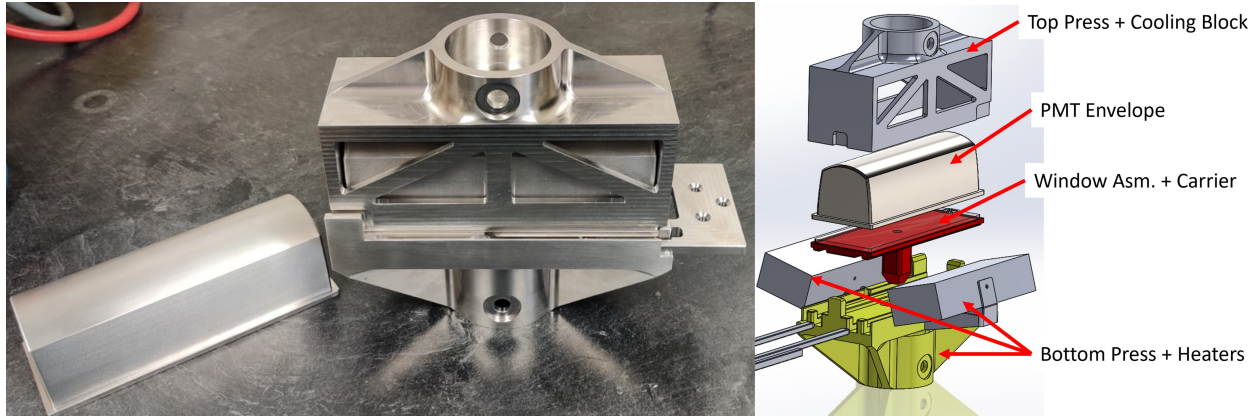


Figure 5.18: *The fabricated assembly-stage components before polishing and lamp mounting.*

External Press The exterior press assembly, much like the interior, was heavily reinforced to ensure component alignment during the assembly process. A force-controlled, Exlar Tritex II linear actuator can apply up to 9,000 N over a 150 mm stroke. An Omega pancake-style load cell is used to sense up to 9,000 N. Custom linear-guided edge-welded bellows were installed to provide transverse rigidity throughout the press stroke, while a steel frame was built to contain the force produced by the linear actuator. Adjustment of the press along the chamber axis is quite simple, achieved by moving the full external assembly. Transverse adjustment is achieved by sliding sleeves mated to the exterior of the main support tubes at the top and bottom of the assembly. Six nuts are welded to the outer tube and bolts can be tightened to affix the outer sliding tube to the inner. A gimbal assembly above the chamber is used to align the stationary, upper-interior press component to the bottom, vertically-moving component. Even pressure must be applied around the photocathode and window perimeter to achieve a quality hermetic seal.

Force Control Application Electronics and control systems for the linear actuator and strain gauge were provided by the Electronics Design Laboratory (EDL) at KSU. LabView was used to construct the control program and user interface, which can be seen in Fig. 5.20 below. Users may provide either positional or force-based control parameters which are then carried out by the actuator. Ultimately, the LabView program proved to be too slow to achieve force-based control. The software provided by Tritex was used instead; however,

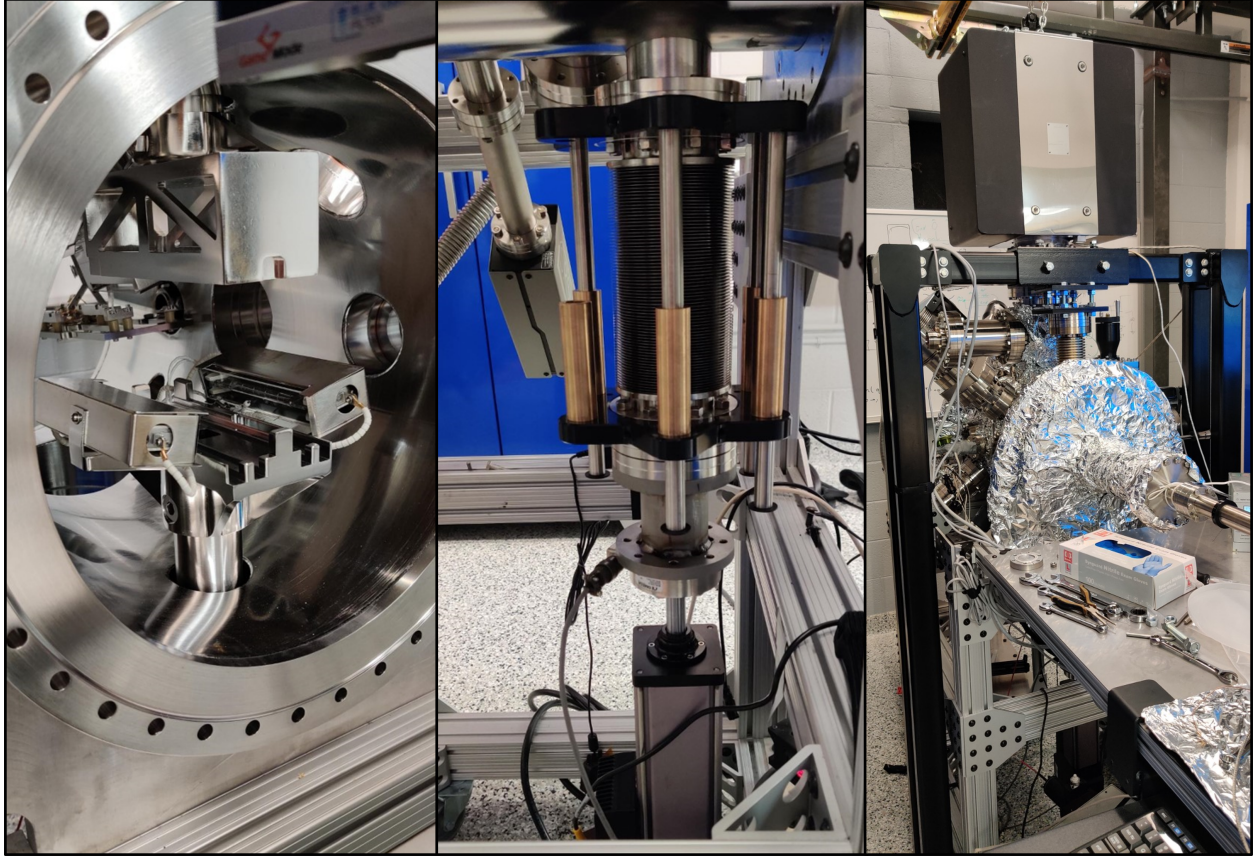


Figure 5.19: *The primary components of the assembly station. Left) The interior press components and lamp heaters. Middle) The press actuator, load cell, and bellows. Right) The external press alignment and force-containment assembly.*

the LabView program is still necessary to monitor and record the applied force.

5.2.5 Process Flow

Figure 5.21 provides a simplified cutaway view of the chamber interior. To process a PMT, the window assembly has a pre-cut indium seal placed on the metalized surface. Small pieces of indium are placed onto the electrical contacts. The window and window carrier are loaded into the antechamber which is then evacuated. The primary valve is then opened and the axial translator is extended to the antechamber, and the magnetic coupler is rotated to lift the window and carrier out of the load-lock, and the valve is closed. The assembly is then moved directly to assembly area and placed on the lower press component. The PMT envelope and dynodes with the dynode carrier are placed in the load-lock. After

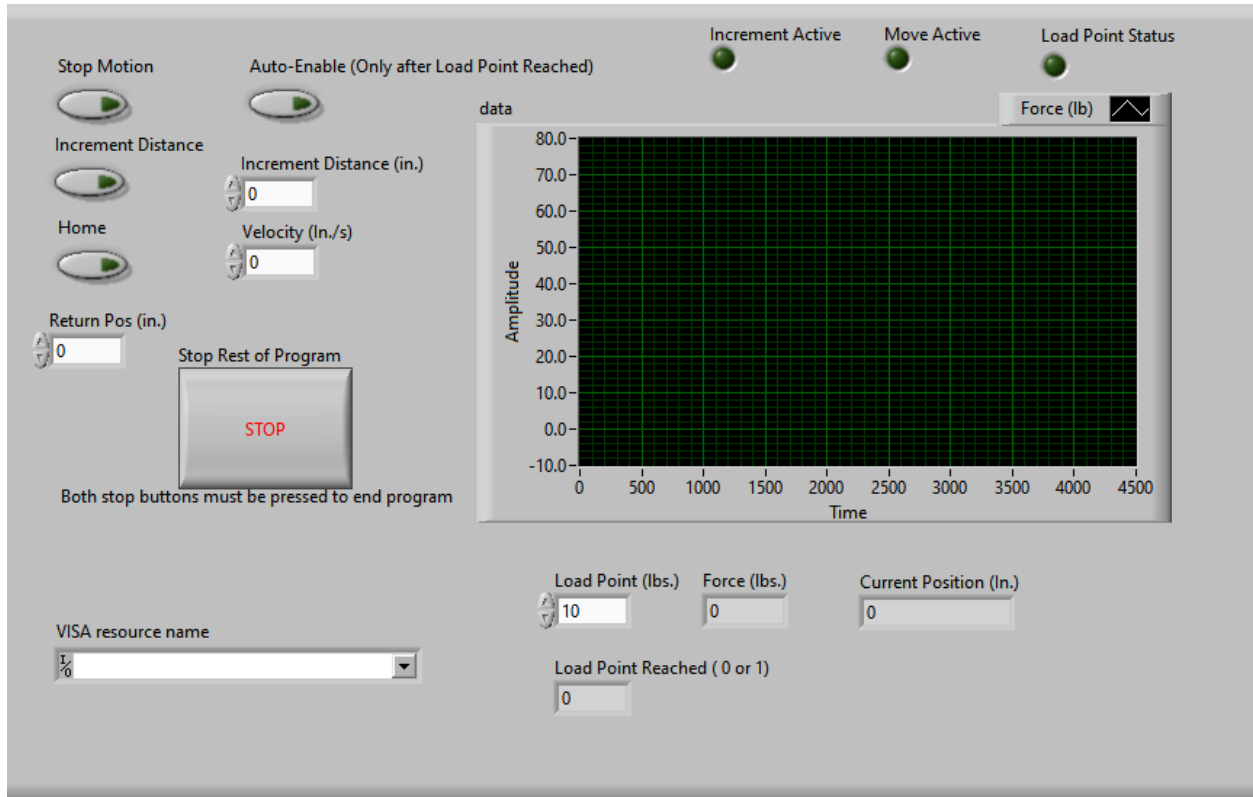


Figure 5.20: *The LabView program used to monitor indium clamping force.*

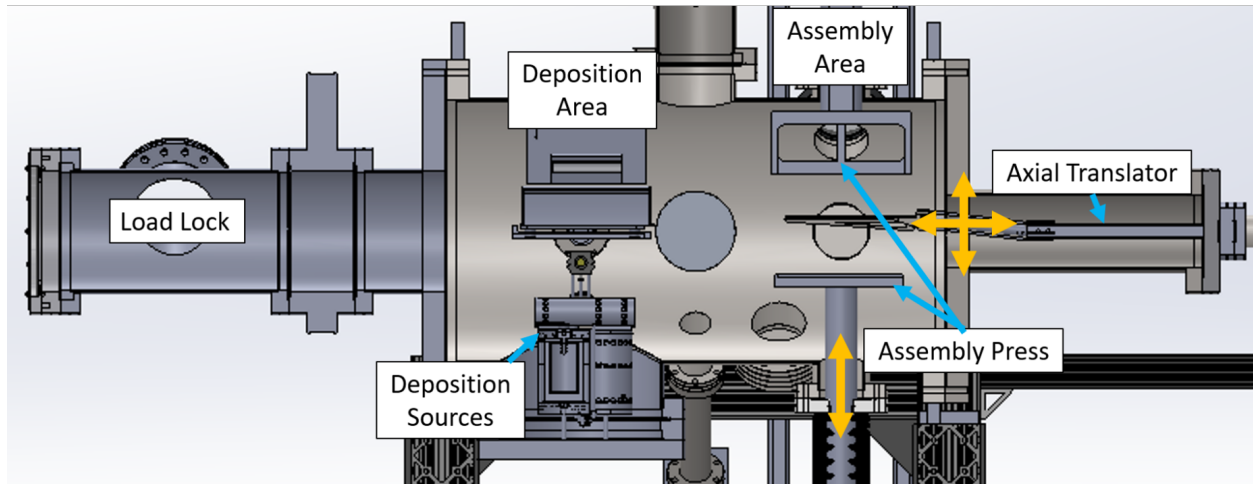


Figure 5.21: *A simplified model of the chamber interior.*

evacuating the antechamber, the valve is again opened and the dynodes are moved by the axial manipulator to the deposition stage. Finally, the PMT is moved to the dynode stage. After a final degassing, the dynodes and PMT are coated with the initial Sb film. Once the Sb source is cooled and vacuum has returned to nominal levels, the alkali deposition is carried

out for each component. After the photocathode and secondary emissive films are grown, the dynodes are taken to the assembly area and are removed from the holder and placed into position by the wobble stick, guided by the tabs designed into the thin window frame. The dynode holder is then placed back in the deposition stage. The axial manipulator is used to move the PMT envelope back to the assembly area where it is placed over the dynodes. Heat lamps are used to bring the indium seal to 120 °C, below the indium melt-point of 157 °C. The lower press assembly and PMT components are raised up to the upper press components, and 4,500 N of compressive force is applied. The integrated substrate cooler is used to maintain a low cathode temperature, so that the photocathode does not degrade due to thermal migration from the heated envelope walls.

Chapter 6

Photocathode Deposition

6.1 Chamber Preparation

Cleaning The vacuum chamber, PMT components, and other internal ancillary equipment utilize UHV-compatible materials and design techniques to reduce the base pressure of the chamber. Machined metal surfaces were mechanically and electrochemically polished to reduce the total surface area on which contaminants could be adsorbed. Both alkaline (10% Simple Green) and acidic (2% Citranox) detergent solutions were used in an ultrasonic cleaner to remove oils and oxidation from component surfaces. Parts were then rinsed with distilled water, and wiped down with acetone, quickly followed by > 99.5% isopropyl alcohol. Components held outside the chamber for later use were wrapped in oil-free UHV compatible aluminum foil to prevent contamination.

Assembly Each chamber flange uses a CF metal gasket seal. Oxygen-free high thermal conductivity (OFHC) copper gaskets are crushed between two flanges which have precisely-machined knife-edges to imprint the softer copper and form a hermetic, UHV seal. Silver-plated copper gaskets were used on flanges which experience high temperatures more frequently, such as those near the TSPs..

Bakeout To reach the lowest-achievable base pressure, a high temperature bakeout was performed over 2 weeks while pumping at near-UHV (10^{-8} torr). The chamber was wrapped with fiberglass-insulated heat tape. The two primary heaters were modulated using Briskheat PID-controlled power supplies. Other heaters, such as those used for the source-drop, bellows, RGA, and ion pump were controlled using variable AC power supplies. Three to four layers of aluminum foil enveloped the chamber surfaces to distribute heat and to serve as insulation.

The chamber temperature was slowly ramped to 150 °C to drive out water and accelerate offgassing. High temperatures were avoided near sensitive turbopumps and vacuum measurement devices. The internal heat lamps, also controlled by variable AC power supplies, brought the deposition and assembly stages up to 275 °C to quickly remove adsorbed water and other surface contaminants.

Similarly, integrated strip heaters evenly raised the ion pump temperature to 200 °C. Each TSP filament was degassed under high vacuum at 53 A until their operation no longer substantially affected chamber pressure. Throughout this process vacuum pressure increased rapidly until it slowly returned to nominal values. While degassing one of the TSPs, hydrocarbons (> 40 AMU) were observed in the RGA spectrum. These contaminants were attributed to previous contamination from a failed cathode growth. This TSP was not used during deposition and did not appreciably affect the vacuum system during depositions.

To reduce the gas load during deposition, the linear effusion cell sources were baked before and after material was loaded. The initial bakeout brought the sources to 350 °C, with the temperature increasing at 10 °C/min. Heat was held at 350 °C for 4 days, while the chamber pressure dropped from 10^{-7} - 10^{-8} torr. After the initial effusion cell bakeout, 99.9999% Sb shot was loaded into the source crucible and outgassed at 350 °C for 5 days to drive out moisture without material evaporation. Both alkali sources and fill tubes were baked at 300 °C for 5 days, as well. A sample RGA spectrum taken during Sb source bakeout can be seen in Fig. [6.1](#).

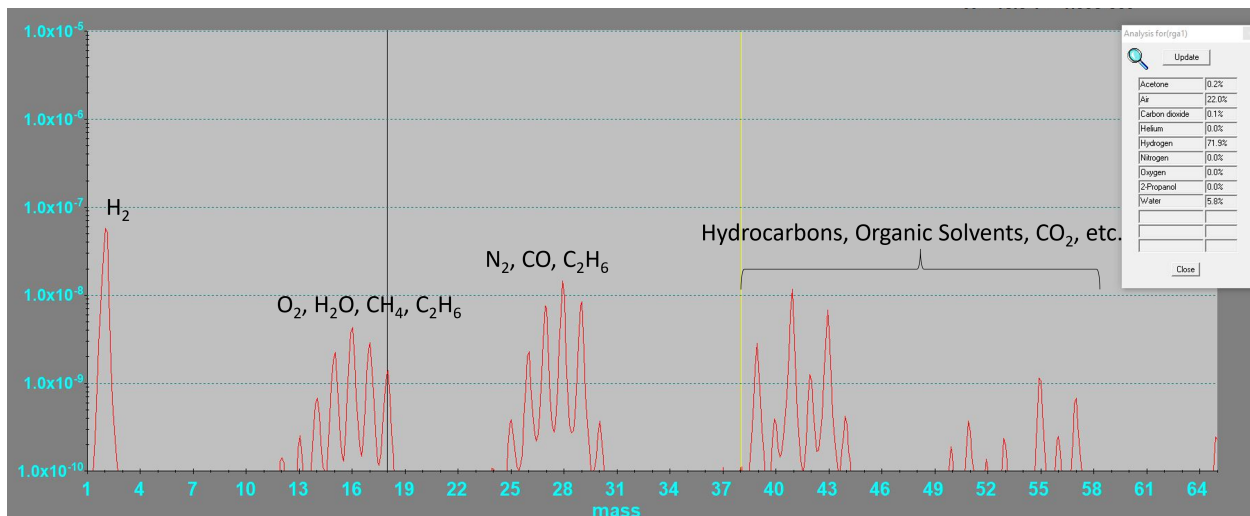


Figure 6.1: *The residual gas spectrum during Sb source and chamber bakeout. Total pressure was 8.9×10^{-8} torr.*

Alkali Source Loading The first deposition attempts used a liquid source transfer process as recommended by SVT Associates. A custom glovebox (Fig. 6.2) was constructed and mated to the chamber's fill-port flange. Ultra-high purity N_2 gas was flowed through the box as a continuous purge-gas to minimize contaminants. Funnels with Swagelok VCR fittings

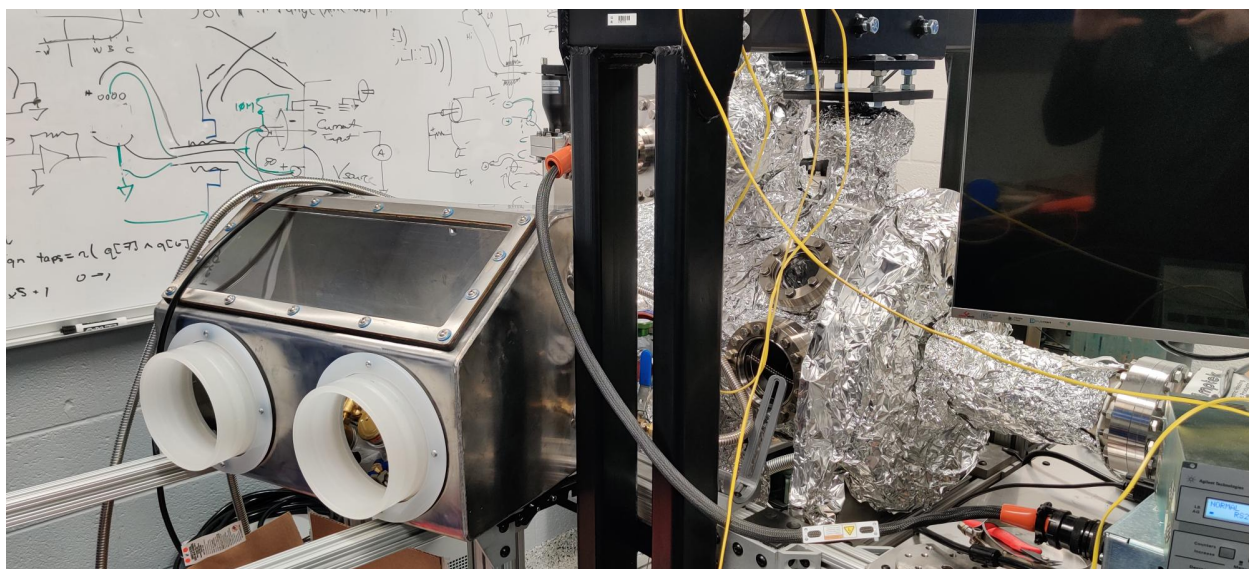


Figure 6.2: *The glovebox used for the first alkali material fill attempts.*

were coupled to the open VCR valves, and the ampules of liquid alkali metals (K and Cs) were poured in. The Cs quickly drained into the heated crucible inside the chamber, however

K, requiring a higher temperature to stay liquid, had seemingly formed a blockage presumed to be caused by insufficient valve temperatures. Therefore, the K fill process was abandoned, and plans were slightly altered to perform Cs_3Sb depositions.

A photocathode was successfully grown — evidenced by qualitative photosensitivity measurements — however, issues interfacing with the Keithley 6487B voltage supply meant that photocurrent could not be accurately quantified. The deposition process was completed, and the power to the effusion cells was removed. The chiller ran continuously, just above freezing, to cool the sources as quickly as possible. Although SVT had claimed that a liquid source transfer process was viable and that the vapor pressure of Cs at room temperature was sufficiently low to prevent evaporation from the effusion cell crucible without valves and shutters, it was disastrously discovered that the source assembly did not have the thermal conductance required to quickly reduce the source temperatures. Upon baking the chamber or operating the Sb source, copious amounts of pure Cs would evaporate and coat the chamber walls. A dangerous, but slow and controlled cleaning process was initially planned; however, one of the cooling-liquid feedthroughs failed during this process, and alkali material was visibly reacting within the vacuum chamber. After a complete chamber refurbishment, it was determined that a vapor-transfer technique should be used such that the alkali sources could always be valved-off in case of equipment failure or other emergency.

For the new source-loading procedure, sealed ampules containing 5 g of 99.95% pure K and 10 g of 99.8% pure Cs from Alfa Aesar were gently placed into individual valved UHV “source containers”, as shown in Fig. 6.3. Each source container mates to Swagelok VCR UHV compatible feedthroughs with two all-metal ball-and-socket valves, bakeable to 300 °C. Resistively heated 12 mm internal-diameter tubing routes the alkali vapor to their respective crucibles inside the chamber. First, the source containers and sealed ampules were connected and opened to the chamber to pump away the remaining atmosphere. The source containers were heated to 200 °C while the chamber maintained UHV. Next, each of the four valves were closed, then separated while maintaining the vacuum of both the sources and chamber. Each holder was shaken to break the fragile glass seal at the end of the ampule. Once the alkali material was visibly draining into the source container, the valves were reconnected

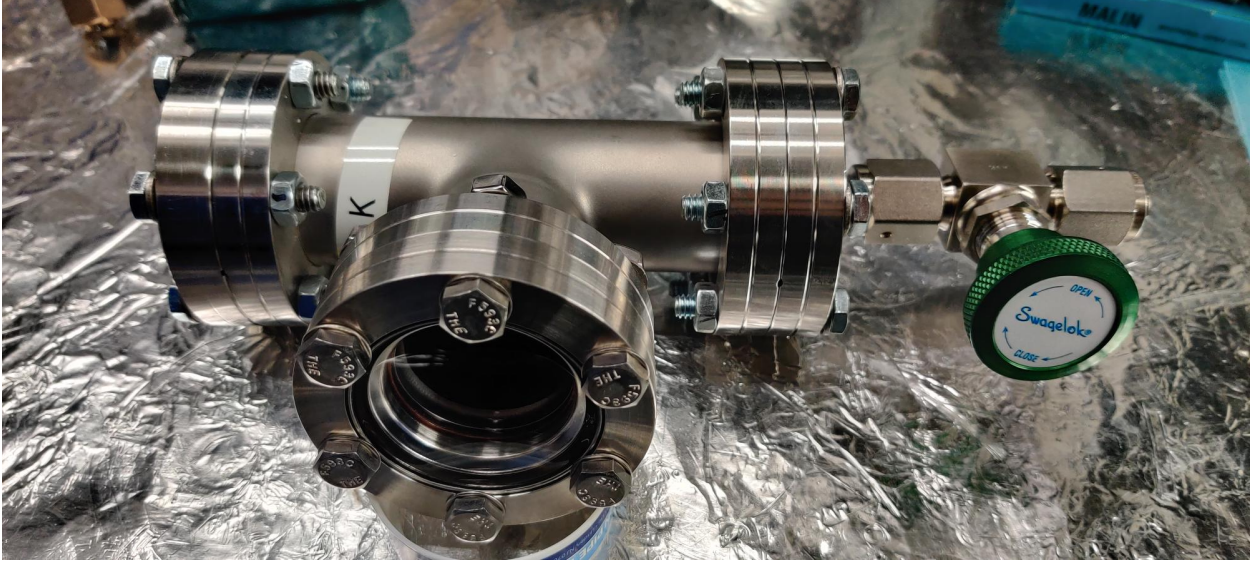


Figure 6.3: *An evacuated ampule containing the alkali material was placed within an evacuated source container and was unsealed with vigorous agitation.*

and the sources were pumped at room temperature.

Alkali Source Testing The total number of atoms required to deposit a material to thickness, d , over area, A , assuming the bulk material density, ρ , is

$$N_{tot} = f_v f_s \frac{d A \rho N_a}{m_a}, \quad (6.1)$$

where f_v is the fraction of atoms impinging the substrate, f_s is the fraction of atoms which “stick” to the substrate, m_a is the atomic mass, and N_a is the Avogadro constant. The atomic flux may be calculated using $\Phi = n_a C$, where n_a is the atomic number density (atoms/l) and C is the conductance (l/s). The atomic number density may be calculated from vapor pressure which is provided over a range of temperatures for a number of materials in the *CRC Handbook of Chemistry and Physics*⁵⁷. Molecular conductance of Cs through a straight tube in the molecular flow regime was estimated following the empirical formula presented by Marquardt¹⁰⁸:

$$C_{mol} = 3.81 \left(\frac{T}{M} \right)^{1/2} \frac{d^3}{l}, \quad (6.2)$$

where T is the temperature in K, d is the inner diameter, and l is the length in cm. The total deposition time is calculated as $t = N_{tot}/\Phi$. Within this equation, a few variables are not easily quantified, and estimated values must be used. The empirical conductance equation is for a straight, uniform tube. Considering the tortuous path through the valve, fill tube and source crucible, it may not be appropriate to use the 45 mm tube length. Similarly, the view and sticking factors for the alkali materials are not well known. The total deposition time at a given temperature is linearly proportional to $l/(f_v f_s)$. In Fig. 6.4, the time required to deposit 110 nm of Cs is plotted against source-holder temperature for a range of values with the coarse assumption that elemental Cs is evaporated as a monomer. Initial testing

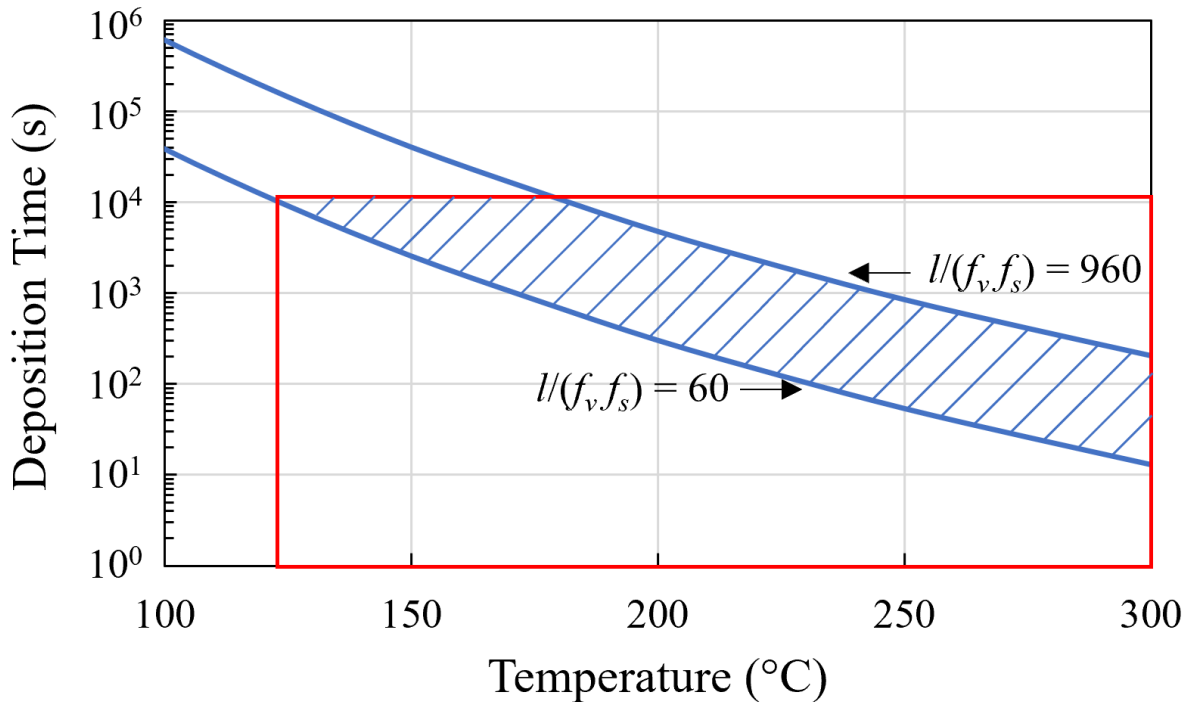


Figure 6.4: The time required to deposit 110 nm of Cs for a range of source temperatures was calculated using the empirical conductance equation. The deposition time at a given temperature is linearly proportional to $l/(f_v f_s)$, which is estimated to provide upper and lower bounds for the expected deposition time. A red box indicates the source temperatures required to coat the substrate in under 3 hr.

at source-holder temperatures up to 300 °C revealed a significant restriction attributed to the dual-valve setup. One of the valves was removed to alleviate this issue, even though the source container and chamber could no longer be separated without repressurizing the

chamber. Further testing up to 150 °C after the removal of a single VCR valve ensured that the source indeed made its way into the chamber, however problems related to material diffusion and flux control persisted, as discussed in later sections.

6.2 Alkali-Antimonide Photocathode Deposition

Alkali-antimonide photocathodes are composed of three alkali atoms for every antimony atom. The standard Cs_3Sb , green-enhanced Rb_2CsSb , and high-performance blue K_2CsSb photocathodes all belong to this family. Development of these cathodes has been ongoing since the 1930s, with major performance gains coming within the last 15 years, largely due to the efforts of scientists developing cathodes for accelerators and phototubes^{63;114–116}. Due to the complexities of photocathode growth, it was decided that initial efforts would focus on a simpler binary compound, thus the following section focuses on the development and growth of Cs_3Sb cathodes. The precursor to K_2CsSb , Cs_3Sb was discovered by Görlich in 1936¹¹⁷. The compound had a much greater photoresponse compared other cathodes of the time. Further investigations of the material’s electrical properties found that the alloy material was indeed a semiconductor, facilitating an extended electron MFP and, thus, higher QE.

6.2.1 Deposition Process Outline

Vacuum Requirements Alkali-antimonide photocathodes are extremely sensitive to contaminants such as O_2 , CO_2 , CO , and H_2O . The growth environment is typically held at total pressures below 10^{-9} torr, with partial pressures of oxygen and water below 5×10^{-11} torr^{118;119}. To achieve these pressures, the chamber and substrate must be cleaned and vacuum-baked to temperatures above 150°C to drive out moisture. Bakeout temperatures $> 300^\circ\text{C}$ are preferred, although vacuum componentry (valves, gauges, pumps) often make this difficult⁵⁴. Similarly, deposition source components and materials previously exposed to atmosphere must be degassed to prevent film and substrate contamination during deposition. Specialty vacuum pumps like ion pumps, TSPs, and NEGAs are used alongside turbopumps

and roughing pumps to achieve UHV. In many cases, the turbopumps may be valved off once sufficient vacuum is achieved to prevent backstreaming, however modern, high compression ratio pumps achieve pressures below 10^{-9} torr which may sustain cathode growth depending on the residual gasses present and the exposure time^{115;120}.

Sb Deposition Historically, Cs_3Sb growth has followed a simple two-step, sequential deposition process. An initial Sb film is deposited and Cs vapor is then introduced while the substrate is held at an elevated temperature. The antimony deposition is vital to the final photocathode response uniformity. It is common to use pure Sb pellets or PtSb alloy for thermal evaporation. Elemental Sb evaporates in UHV at temperatures $\gtrsim 450$ °C, and antimony is liberated from PtSb at $\gtrsim 1000$ °C¹²¹. While evaporating antimony onto a glass substrate, a semitransparent, amorphous brown film will develop. Once a critical thickness is attained, small, opaque patches will begin to form. The patches will spread and merge to form a homogeneous film with a metallic sheen. This coalescence is associated with crystallization, and the coalescence thickness varies with the source type, evaporation rate, and substrate temperature. Pure Sb evaporation tends to produce clustered Sb_4 , whereas PtSb sources are more likely to emit monoatomic Sb. Clustered Sb_4 tends to form nucleation sites and produces a lower-density, rougher film with higher resistivity¹²². The crystallization thickness is reduced at higher substrate temperatures as the atomic Sb is more mobile on the substrate surface. Target substrates are typically held between 100-160°C during Sb deposition^{118-120;123}.

To measure the thickness of an antimony film, either a QCM or optical-transmission measurements may be used, substrate permitting. The preferred thickness or measured transmission is usually found through process optimization, since the deposition material, rate, and substrate affect the final material properties. The thickness of the Sb film is often application-specific. Photon absorption may be tuned to specific wavelengths by modifying process parameters such as the film thickness¹²⁴. The crystallization begins at a thickness of approximately 4 nm¹¹⁹ and continues until a thickness of 15-25 nm¹²², which is thicker than a common semitransparent photocathode³⁴ but generally on the order of a reflection-type

cathode. For phototubes, the optimal light transmission through an Sb film from a tungsten lamp is typically 70-85%, which correlates to a thickness of 4-6 nm^{34;121}.

Cs Deposition Photocathode synthesis is achieved by slowly introducing cesium vapor to the antimony film. Alkali materials are difficult to handle, and pure materials cannot be exposed to a typical atmosphere. To simplify the deposition process, sealed or compound-material, capsule-style sources from Alvatec or SAES may be used. These sources have a relatively low material yield, and are best served as single-use sources. Alternatively, pure materials or chromate sources mixed with a getter material, such as Ti, may be used. During Cs deposition, the substrate temperature is held between 90-150°C, depending on the recipe followed^{34;118;120;123}. The elevated temperature promotes Cs diffusion into the Sb, however a maximum temperature near 200°C will begin to decompose the compound³⁴. Ultimately, the substrate temperature is chosen to balance Cs diffusion into the material while limiting Cs rejection at the surface. Cathode performance is monitored *in situ* during Cs deposition. The photocurrent emitted from the cathode slowly rises until it plateaus at the optimal thickness. Continued Cs deposition will eventually reduce the QE, though excess material may be removed by heating. A rise in QE is typically seen as the substrate cools, since fewer thermally excited electrons occupy the conduction band thereby reducing the frequency of electron-electron scattering. The final film thickness is typically 8× the thickness of the original Sb layer¹²⁵. For bialkali films, it has been shown that Cs enhances QE beyond simply lowering the band gap. In a sequential bialkali growth, the relatively weak K₃Sb bonds are broken to include Cs. It is believed that the addition of Cs enhances the crystallinity, thereby reducing defect scattering and increasing the electron MFP. A superficial layer of oxygen has been shown to further improve photoemission up to an additional 50% through band bending^{117;118;126}. The evolution of photoemission during Cs deposition can be seen in Fig. 6.5. Alternative growth methods, such as simultaneous co-deposition and sputtering have been performed^{116;127}. Codeposition of bialkali cathodes have shown enhanced the photoemissive properties^{63;127}, however it is quite difficult without actively monitoring stoichiometry.

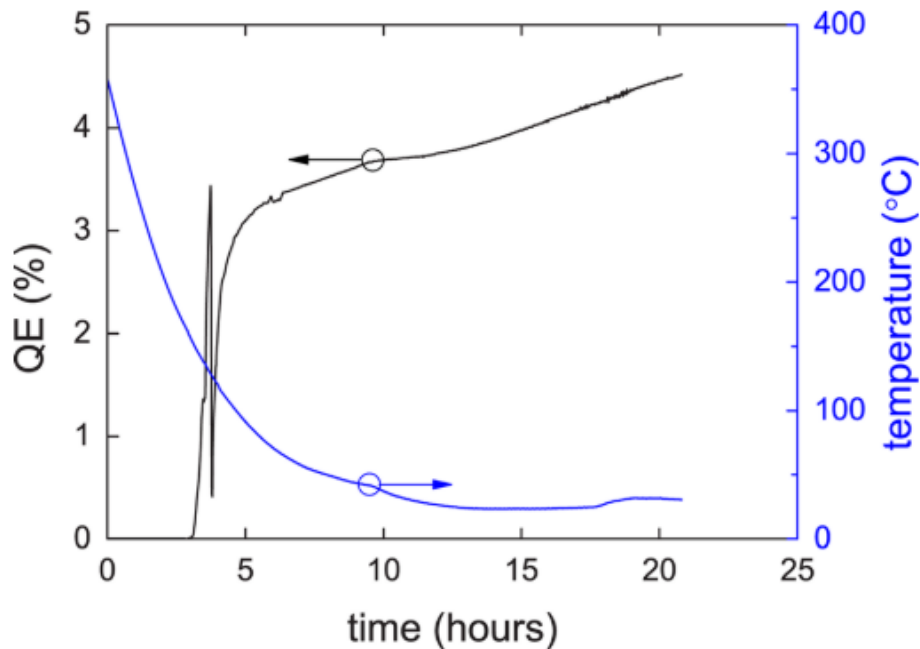


Figure 6.5: *The QE (at 532 nm) evolution of an SbCs photocathode during Cs deposition performed at Cornell³¹.*

6.3 Alkali Antimonide Deposition Attempts

6.3.1 Substrate and Chamber Preparation

A chemically cleaned PMT envelope/cathode substrate was placed into the chamber on the deposition stage. A bakeout procedure brought the substrate to 250 °C for one week, while the chamber was held at 150 °C, and the internal sources at 300-350 °C. The chamber pressure did not fall below 6×10^{-9} torr with excess amounts of air and water present in the RGA spectrum. A search found that the the uncapped Cs fill-port valve had a small leak which was subsequently sealed. During a calibration deposition to find the tooling factor for the QCM, another air leak arose, and the thermal deposition sources appeared to short-circuit at high temperatures. Disassembling the source flange revealed an oxidized copper gasket due to high source temperatures. Further disassembly of the Sb source crucible revealed that antimony and evaporated and condensed on the ceramic heater-wire insulators which had made contact with the thermal shielding. Both issues were rectified, and the chamber was reassembled with a silver-plated gasket at the source drop. A two-week bakeout brought the

chamber to its base pressure of 7×10^{-10} torr, as seen in Fig. 6.6. A partial leak-down test provided a leak-rate of 3.73×10^{-9} torr l/s from the combined leak and outgassing sources.

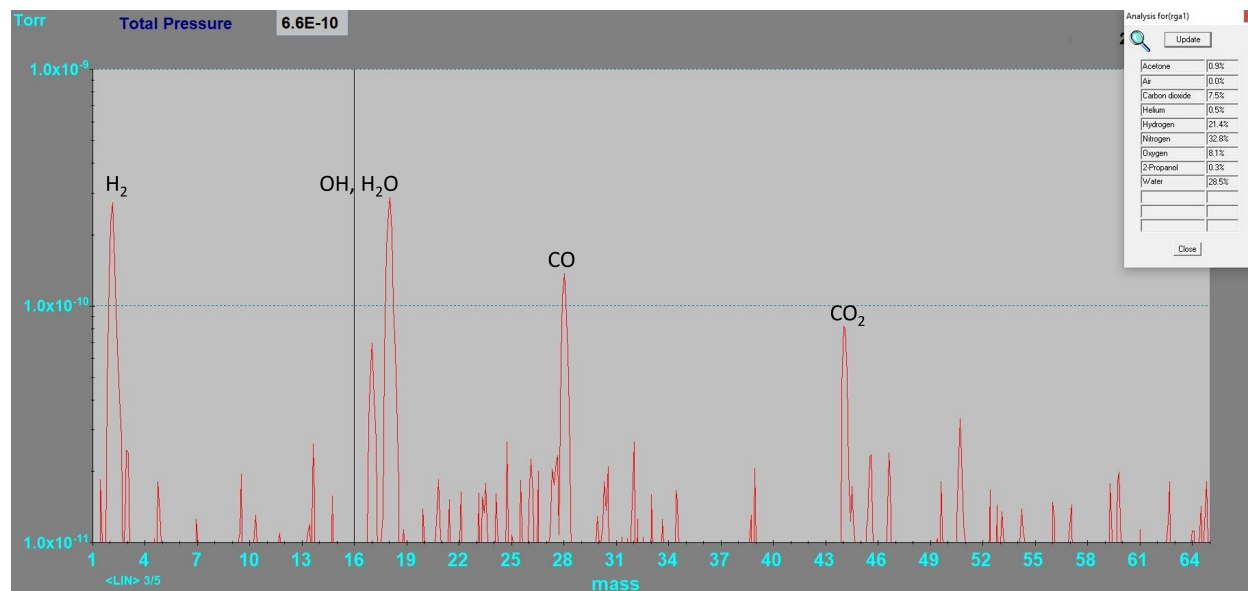


Figure 6.6: *The residual gas spectrum after baking the chamber. The mass spectrum shows excessive water, which is otherwise getterred by operating the TSP.*

6.3.2 Photocathode Deposition

For the antimony deposition, the cathode temperature was reduced to 120 °C. The Sb effusion cell crucible temperature was increased to 530 °C, while the manifold was heated to 550 °C to prevent condensation. Prior to a typical Sb evaporation, with the source near deposition temperature, the total pressure was $\sim 7 \times 10^{-9}$ torr, with partial pressures of O_2 and H_2O below 10^{-9} torr. To begin, the QCM shutter was opened and monitored until the deposition rate reached 0.01 nm s^{-1} . The substrate was translated to the Sb deposition location, and the evaporation proceeded at a rate of $0.015\text{-}0.05 \text{ nm s}^{-1}$ until 10-60 nm (depending on the run) of Sb was deposited as measured by the calibrated QCM (Fig. 6.7). During Sb deposition, total pressure would rise to $\sim 2 \times 10^{-8}$ torr, while the partial pressures of O_2 and H_2O remained below 10^{-9} torr. The increased pressure was primarily attributed to additional H_2 and CO from the Sb source.

Following Sb deposition, the Sb source was allowed to cool for 3-4 hours while the chamber

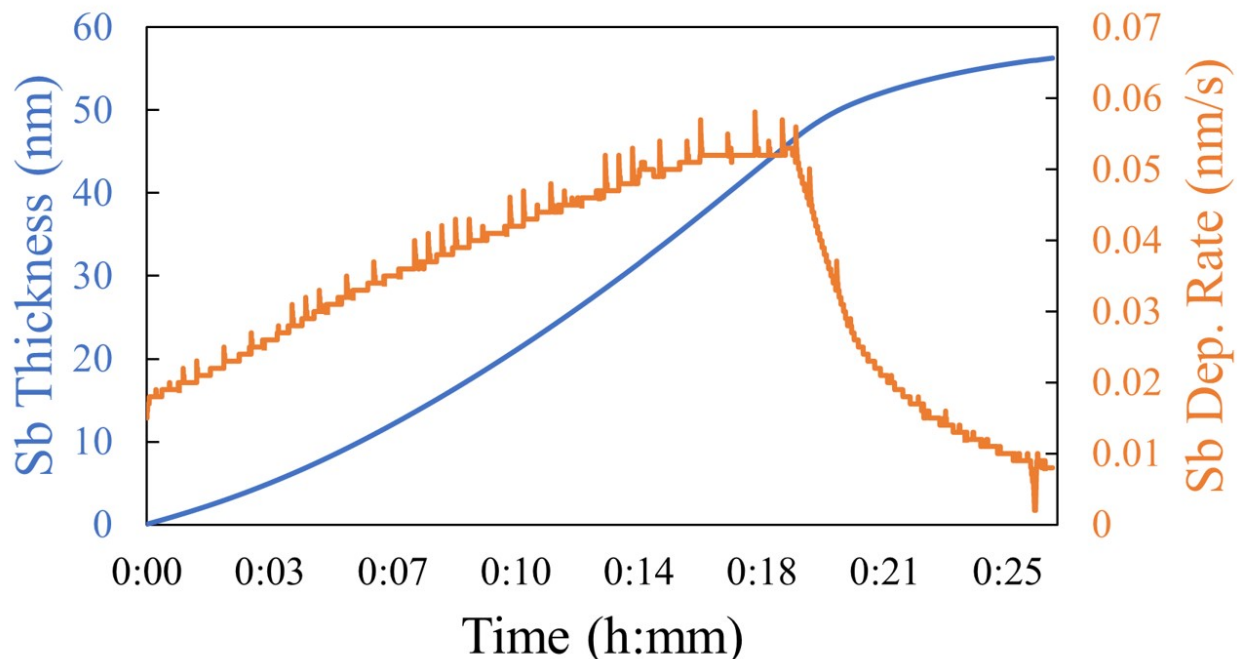


Figure 6.7: *Growth rate and thickness vs. time for the antimony deposition on 9/14/2023.*

pumped down to $\sim 10^{-9}$ torr. Depending on the run, the cathode substrate was held between 90-140 °C. The external cesium source and fill tube were heated between 125-200 °C as measured by a type-K thermocouple and compatible digital multimeter. The effusion cell crucible was brought to ~ 115 °C and the manifold was held at ~ 135 °C. Prior to the initial Cs source tests, 60 nm of Sb was coated on the cathode substrate to enable photocurrent measurement in the case that Cs_3Sb was produced. As previously stated, it was discovered over the course of 4 hours that the dual-valve system restricted the flow of Cs vapor, even at high source-holder temperatures, due to the complex geometries of the Swagelok VCR valves and effusion sources. To reduce the restriction, the vacuum system was shut down, and the chamber was backfilled with N_2 through the K port. The chamber-side valve was removed, and the source was quickly reinstalled. The antechamber valve was quickly opened to swap to a clean substrate, and the chamber was pumped down for 1.5 weeks. The Cs source was to be opened to the chamber to maintain good vacuum, however upon reintroducing the source to the chamber, an unexpected spike in pressure occurred. It was initially thought that Cs had quickly found its way to the chamber after removing one of the valves, however

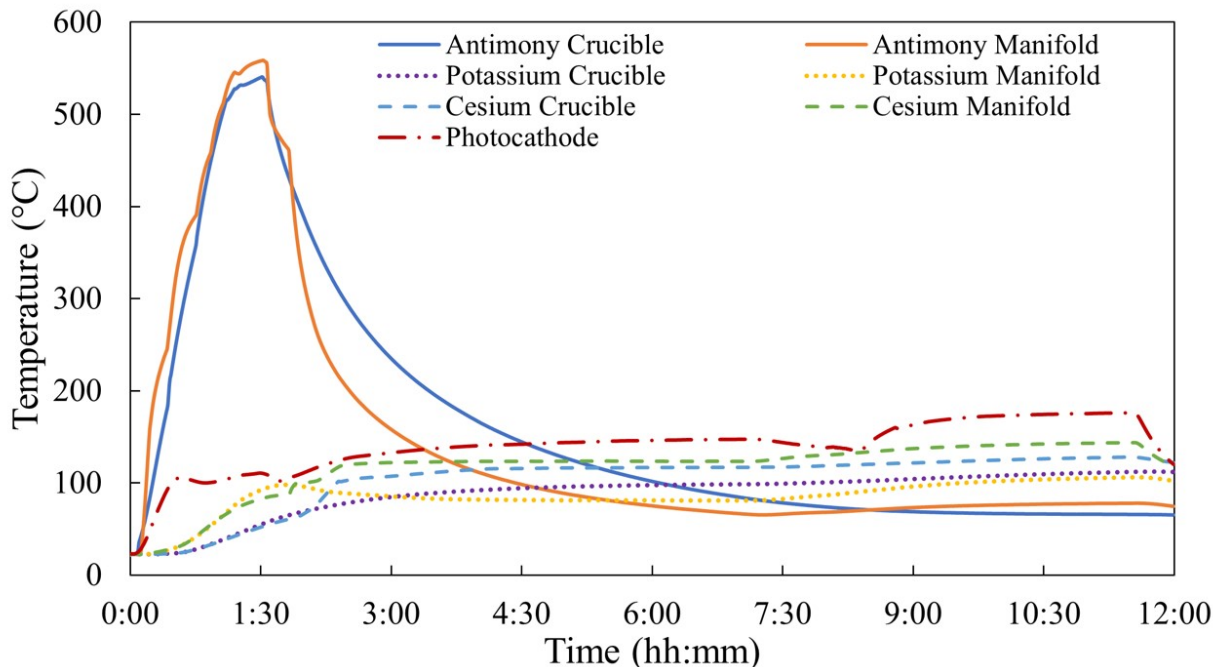


Figure 6.8: *The temperature of each deposition component for Sb and Cs deposition which took place over the course of 8 hours on 9/14/2023.*

the RGA showed an unexplained increase in the primary constituents of air.

With the Cs valve shut, another Sb deposition was performed at a base pressure of 1.2×10^{-9} torr with the partial pressure of H_2O at 1.3×10^{-10} torr. During the deposition, the chamber pressure rose to 2.5×10^{-8} torr, while the partial pressure of water reached 7.4×10^{-9} torr. 58 nm of Sb was deposited with the approximate substrate and source temperatures of the previous deposition. The chamber was allowed to pump down for approximately 1 hr, while the antimony sources cooled. Upon opening the Cs source, the pressure again shot to 10^{-6} torr before quickly settling to 5×10^{-8} torr.

It was noted during this time that the leakage current on the photocathode would rise and fall with the pressure. The picoammeter had otherwise maintained leakage currents of 0.020-0.042 nA, linearly increasing with applied voltage from 200-350 V. This current was relatively stable with all of the vacuum equipment operating under UHV conditions, however further testing in the high-vacuum regime revealed a positive correlation between pressure and leakage current. Pressure was increased by opening the valve to the Cs source which had an apparent, albeit small, air leak. The collection voltage and incident radiant flux were

varied to see if there was a proportional response which could be associated with a simple leakage current or photocurrent, however no significant response (relative to the increased current) was observed. Figure 6.9 provides the annotated picoammeter measurement. After

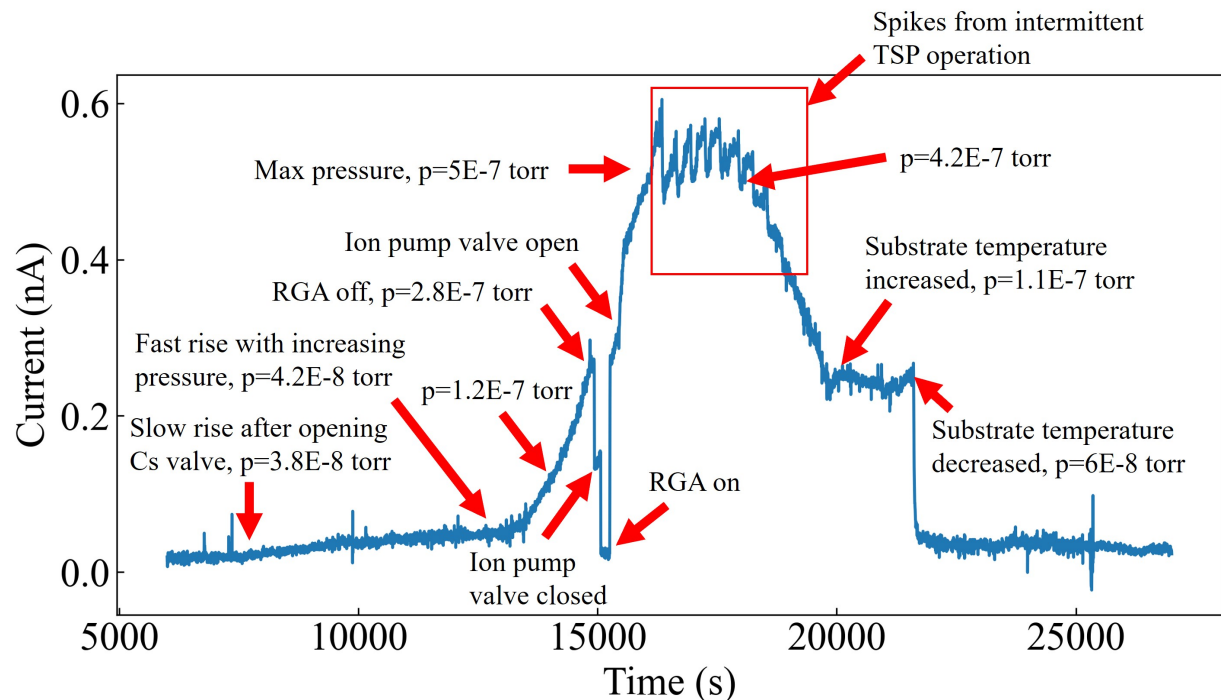


Figure 6.9: Current changes due to vacuum equipment operation with pressure rising in the background. No significant changes in current are observed while operating at a fixed voltage below 10^{-8} torr, regardless of the substrate temperature, RGA operation, or ion pump functionality.

reaching a pressure of 5×10^{-8} torr and reducing the substrate heater power, the current returned to approximately leakage-current levels (0.03 nA at 250 V). Given the elevated response with increased pressure, amplified by the ion pump and RGA, it was determined that ion feedback was the culprit. At typical photocathode deposition pressures, below those which affected the measurements above, the level of ion feedback does not greatly affect photocurrent measurement. Of note is the initial slow rise in pressure followed by a sharp increase, indicating that molecular transport from the source container to the substrate has a substantial delay due to poor conductance. The increase in pressure was frustratingly observed to be highly variable across a range of source-holder, tubing, and effusion cell temperatures.

Furthermore, throughout this process the heated Cs had evaporated and condensed on the relatively cool source-holder viewport. While most of the condensed material had a metallic sheen typical of pure Cs, small, dull patches had formed. It is believed that these were patches of oxide and hydroxide forming due to residual gasses from the apparent source-holder air leak observed in the RGA spectrum. Figure 6.10 shows one such region of reacted Cs. It is likely that the heat applied to the source-holder viewport flange while trying

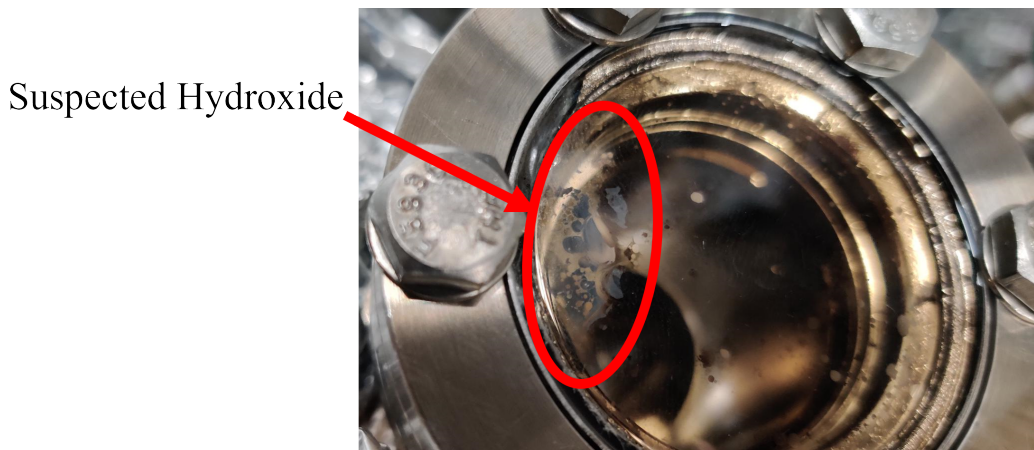


Figure 6.10: *Reacted Cs appears to have formed on the viewport due to a small leak.*

to deposit Cs with the dual-valve setup caused the copper gasket to fail and form a leak. The viewport flange bolts were further tightened, which proved effective when the holder underwent further leak testing.

Attempting another deposition, the substrate was heated to 350 °C to remove any contamination layer and then cooled to the alkali deposition temperature of 90 °C. The vacuum chamber reached 1.2×10^{-9} torr with a water partial pressure of 5×10^{-10} torr, and another Cs deposition was attempted on the existing Sb layer. The internal Cs source components were heated to 130 °C and 145 °C, respectively. The external alkali source was opened to the chamber and the pressure was monitored for any changes over the course of an hour. No change in photocurrent or chamber pressure were observed, as expected. The Cs container was heated to 200 °C over the next 2 hours. Again, an increase in chamber pressure was observed after a delay. The picoammeter current climbed from 0.04 nA at 250 V to 0.2 nA. Total pressure suddenly rose from 6.8×10^{-8} torr to 1.7×10^{-7} torr while water increased

from $9.45 \times 10^{-10} - 3 \times 10^{-9}$ torr, beyond appropriate high quality cathode deposition pressures. The reported ion gauge pressure surpassed 10^{-6} torr in major disagreement with the reported ion pump and RGA pressures, the latter of which was previously calibrated to the ion gauge response. The difference in reading is attributed to alkali vapor condensation on the cold cathode ion gauge, causing a leakage current and rendering it useless for pressure measurement following alkali deposition.

During Cs deposition, the leakage current frequently overwhelmed the picoammeter range. The internal buffer was initially programmed to store, average, and output 25 individual measurements. During each series of measurements, a current-range overflow would invariably render the average value useless. After a time, the program controlling the photocurrent measurement crashed, and values were manually recorded during a spectral scan. These results from September 24th, 2023 are plotted in Fig. 6.14 without the associated measurement uncertainty which could not be tabulated. In response, the deposition was halted, and the photocurrent measurement code was rewritten to reject overflow values — shifting the averaging and uncertainty calculations over to the process control computer enabling overflow rejection.

After this deposition, doubly ionized Cs appeared in the RGA spectrum. The ratio of doubly-ionized Cs to singly ionized Cs is unknown due to the limited RGA mass range. However, the first ionization energy is 3.89 eV, while the second ionization energy is far higher at 23.16 eV⁵⁷, meaning that the total partial pressure of Cs may be one to two orders of magnitude higher than that of doubly-ionized Cs alone. Doubly ionized Cs can be seen in the RGA spectrum shown in Fig. 6.11. The partial pressure of Cs^{++} is approximately 3×10^{-10} torr, with the highest contribution to total pressure being H_2 at 9×10^{-9} torr. The total pressure reading (4.6×10^{-8} torr) is far higher than the sum of the contributions of the other constituents implying a large partial pressure of an unknown source assumed to be entirely Cs. Furthermore, the chamber viewports had attained a yellow tint similar to that of a PMT photocathode suggesting highly diffuse emission of Cs. An excess of H_2 is expected in UHV systems at low pressure due to difficulty pumping small contaminants; however, analyzing the spectrum suggests that residual H_2 , O_2 , and OH may stem from the

dissociation of CsOH at high temperatures. The presence of these molecules confirmed the earlier supposition that an air leak had contaminated the pure Cs source.

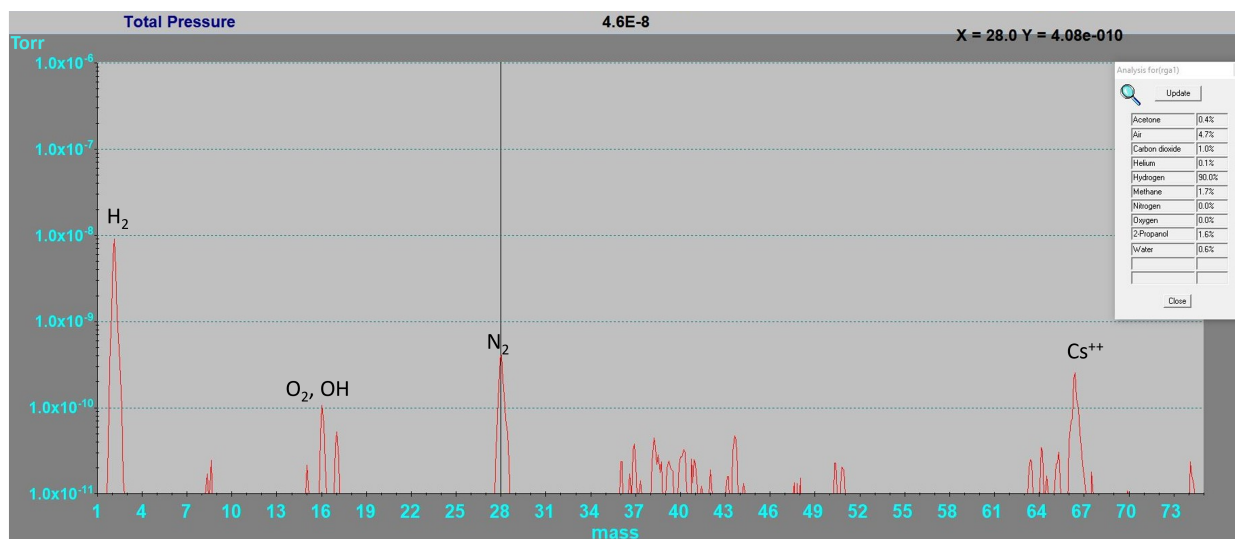


Figure 6.11: *The vacuum chamber environment after Cs deposition. The addition of Cs⁺⁺ to the residual gas spectrum suggests a much larger amount of Cs coats the chamber interior.*

Another 10 day bakeout was performed to remove much of the excess Cs and recover the base pressure. Early in the bakeout, Cs evaporated from the chamber walls and cathode substrate and began to condense on the insulators which maintained discontinuity between the chamber and photocurrent measurement system. A leakage current of 1.5 μA was measured at 150 V collection potential. The substrate temperature was further increased to 300 $^{\circ}\text{C}$ which effectively removed the condensed Cs and brought the leakage current down to approximately 350 nA.

A final 10 nm Sb deposition was performed under a 1.2×10^{-8} torr vacuum at 0.02-0.04 nm/s on October 3rd. The Sb source was allowed to cool and the Cs sources were brought to 150 $^{\circ}\text{C}$. Opening the source valve, the pressure quickly rose and then subsided to 5×10^{-8} torr. The current similarly rose and subsided to 1.3 μA , which was confirmed to be purely leakage. The valve was left open over the course of an hour. Leakage current again spiked and fell to approximately 2 μA . The valve was closed, and the current was continuously monitored using the relatively low-powered broadband source. Intermittent photosensitivity testing was performed using a flashlight to attain a better signal-to-noise

ratio. A screenshot of the photocurrent monitor GUI with a photocurrent response taken on October 3rd, 2023 may be seen in Fig. 6.12. The 275 nm source was connected to measure

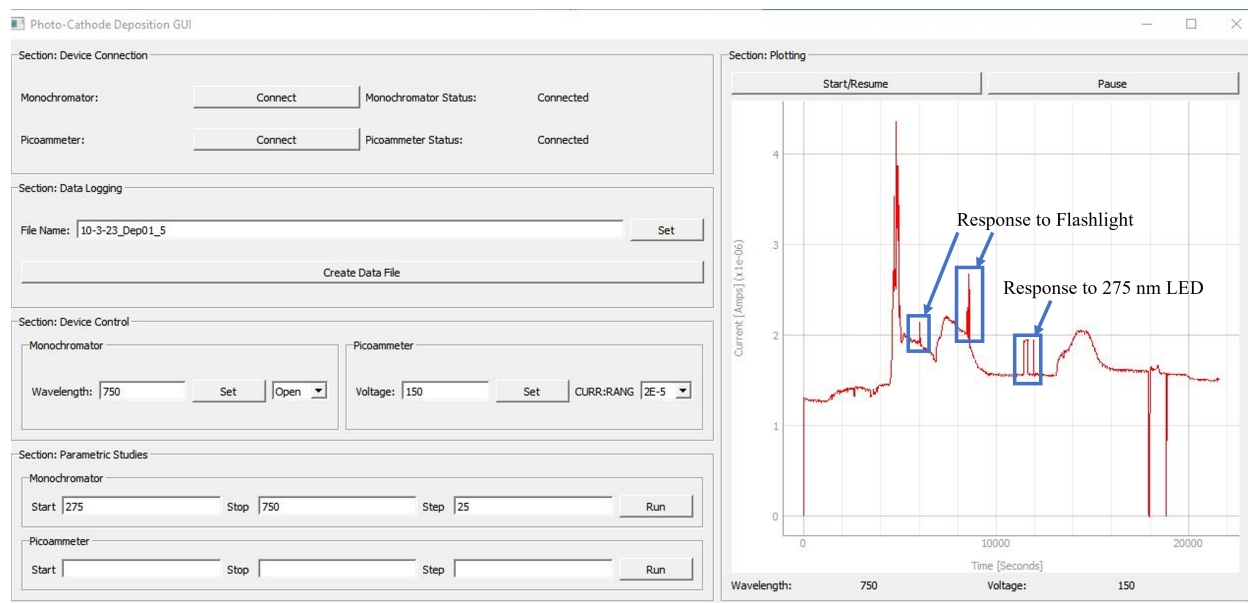


Figure 6.12: The custom GUI used to monitor photocurrent evolution throughout the alkali deposition process. A deposition performed on October 3rd, 2023 was live-plotted and may be seen in the photocurrent measurement window.

photocurrent from a stronger calibrated source. The response to the 275 nm light may also be seen in Fig. 6.12. Following these measurements, the broadband source was reconnected and spectral measurements were taken over the dynamic background, as seen in both Fig. 6.12 (18,000-18,500 seconds) and the extracted data in Fig. 6.13.

6.3.3 Deposition Results

The wavelength-correlated data for two depositions may be seen in Fig. 6.14. Both photocathodes exhibited poor quantum efficiency across the full spectral range. Parzyck *et al.*¹²⁸ found that Cs-deficient CsSb cathodes are less efficient photoemitters than their stoichiometric counterparts and have a much higher emission threshold of ~ 2.19 eV (566 nm) compared to 1.65 eV for Cs₃Sb. With the caveat of high uncertainty at these wavelengths for the presented cathodes, this threshold energy lies near those shown in Fig. 6.14. It was also found in the same study that the Cs-deficient cathodes were more chemically robust¹²⁸. Given the

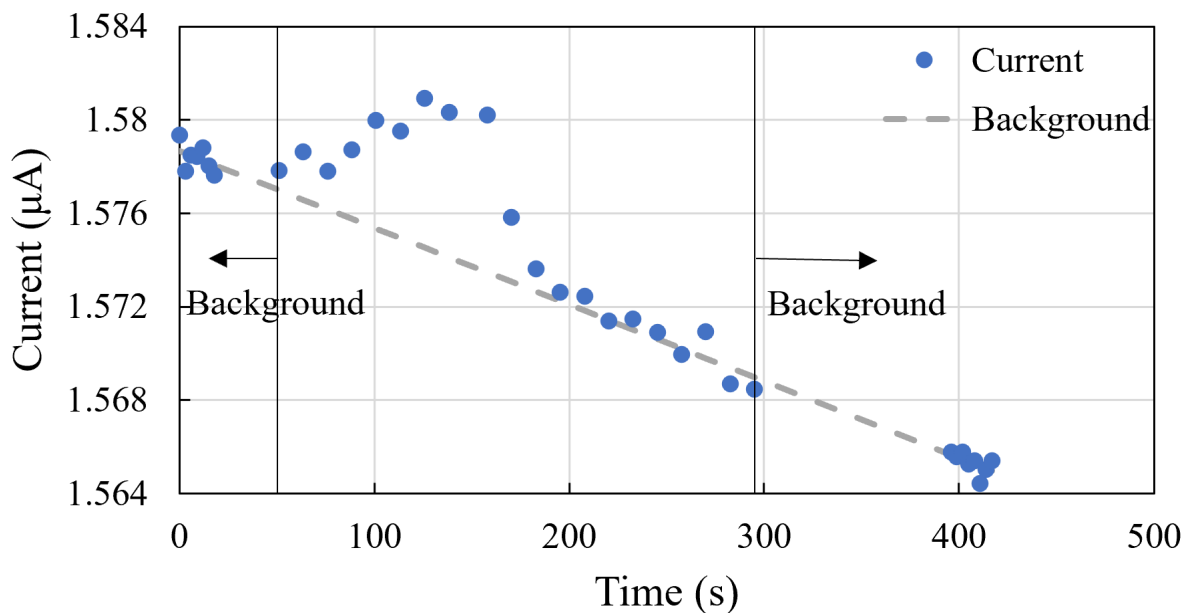


Figure 6.13: *Spectral QE measurements over the large and dynamic leakage background current.*

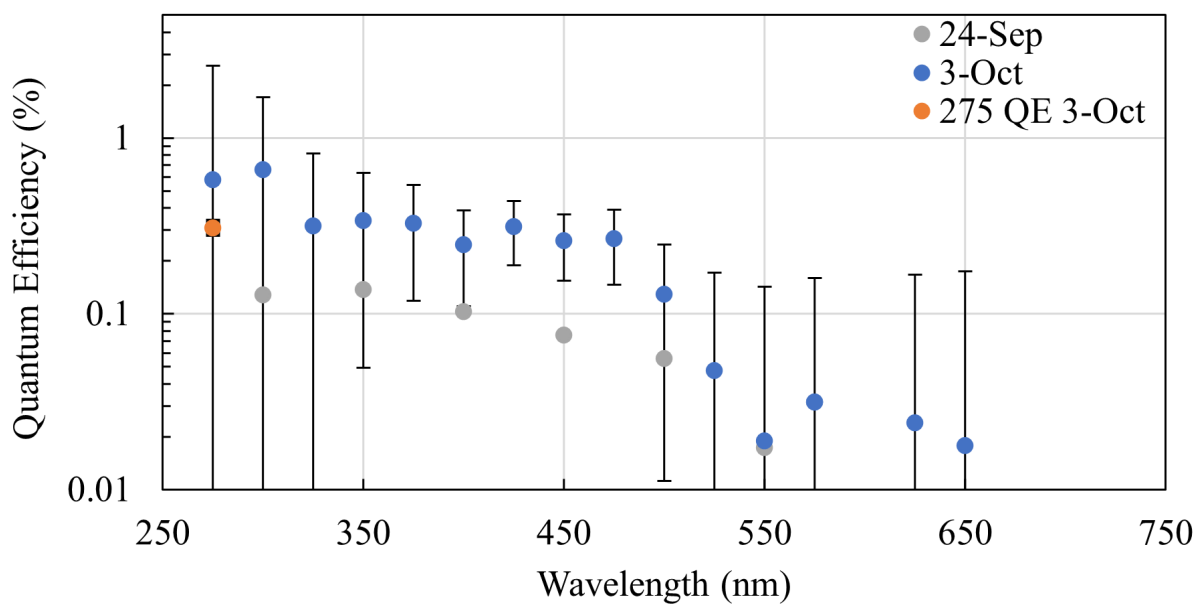


Figure 6.14: *Quantum efficiency of two cathodes grown on September 24th and October 3rd. Leakage current as a result of Cs deposition on insulating materials made precise measurement difficult.*

relatively poor growth environment, contaminated Cs source, and uncontrolled flux leading to shortened deposition times, it is likely that insufficient Cs diffused into the thick Sb layer. For future depositions, it is recommended, at the very least, that the contaminated Cs source is substituted for fresh material. Additional changes to deposition and measurement componentry which will improve the deposition process are outlined below.

Proposed Component Modifications Improvements to the current system may be achieved by modifying a number of existing components.

1. The vacuum may easily be improved by replacing the contaminated TSP elements. Alternatively, a simple swap of the ion-pump mounted TSP to the contaminated TSP location would immediately improve the pump-rate.
2. Ceramic and polyimide insulators may be shielded from Cs deposition by cleverly-positioned sheet metal and UHV foils.
3. The source manifold would be better adapted to alkali deposition with a tighter emission distribution. The current hole distribution, designed to uniformly coat Sb over a large-area substrate, disperses the already diffusely-emitted alkali material throughout the chamber, negatively affecting the vacuum and measurement equipment.

Still, the apparent limitations of the current chamber and effusion cell designs will continue to negatively impact photocathode growth and lifetimes. The large volume and expansive surface area exposed to vacuum flood the chamber with outgassed contaminants when heated, and the poorly-controlled alkali flux further overwhelms the pumps. The large thermal mass of the effusion sources means that they are difficult to cool in a timely manner and will continue to outgas at a relatively high rate over an extended period. Exposure to subpar chamber vacuum actively degrades cathode quality and slows the sealing process while the vacuum is recovered.

Relatively simple changes to the alkali sources would enable a controlled deposition process and reduce outgassing. In a proposed modification to the current source design, an

alkali ampule would be loaded into a copper or thin-walled stainless steel tube just larger than the diameter of the ampule itself. One end of the tube is permanently sealed, while the other is mated to an all-metal bellow or butterfly valve connected to the chamber. A resistively heated tube replaces the crucible inside the chamber. A sealed ampule is loaded into the external source tube which is then evacuated and baked. To release the alkali material, the external tube is partially crushed, breaking the ampule inside. The large cross section enables high conductance, while the combination of temperature and valve modulation precisely control the alkali flux. An external Y-junction may enable the use of a single manifold, further reducing the vacuum load. Lower temperature processing and fewer vacuum seals mitigate the likelihood of forming a leak. Additionally, repositioning the QCM would maintain quantitative Sb deposition monitoring, while providing a qualitative method of observing alkali deposition, which is now understood to be necessary given the delayed vacuum response after source activation. Although a quantitative measure of alkali deposition is possible in theory, in practice, the disparity between substrate and QCM sticking coefficients combined with the challenge of thickness calibration renders it nearly impossible. Still, there is value in knowing precisely when the alkali material begins to coat the substrate beyond just observing the photocurrent. An illustration of this concept, similarly implemented in other work¹²⁹, is shown in Fig. 6.15.

Proposed Process Modifications The Large Area Picosecond Photodetector (LAPPD) project has highly uniform bi-alkali photocathodes with nearly 30% QE^{32;130}. The deposition process enables simpler atmospheric (or glovebox) assembly of the photodetectors prior to sensitive material deposition. A $22 \times 22 \text{ cm}^2$ window with a predeposited NiCr/Cu layer for In sealing is loaded into a UHV system where an 8-10 nm Sb film is deposited. The coated window is removed from the vacuum deposition chamber and mated to the lower assembly which already contains the multiplier and insulating support structures. The device is transferred to a HV system, and a 300-350 °C bakeout is performed over 24-34 hours to seal the device using a molten indium-capillary sealing process, while removing the thin oxide layer formed at the Sb surface. The tile is cooled to 120 °C, and a leak check

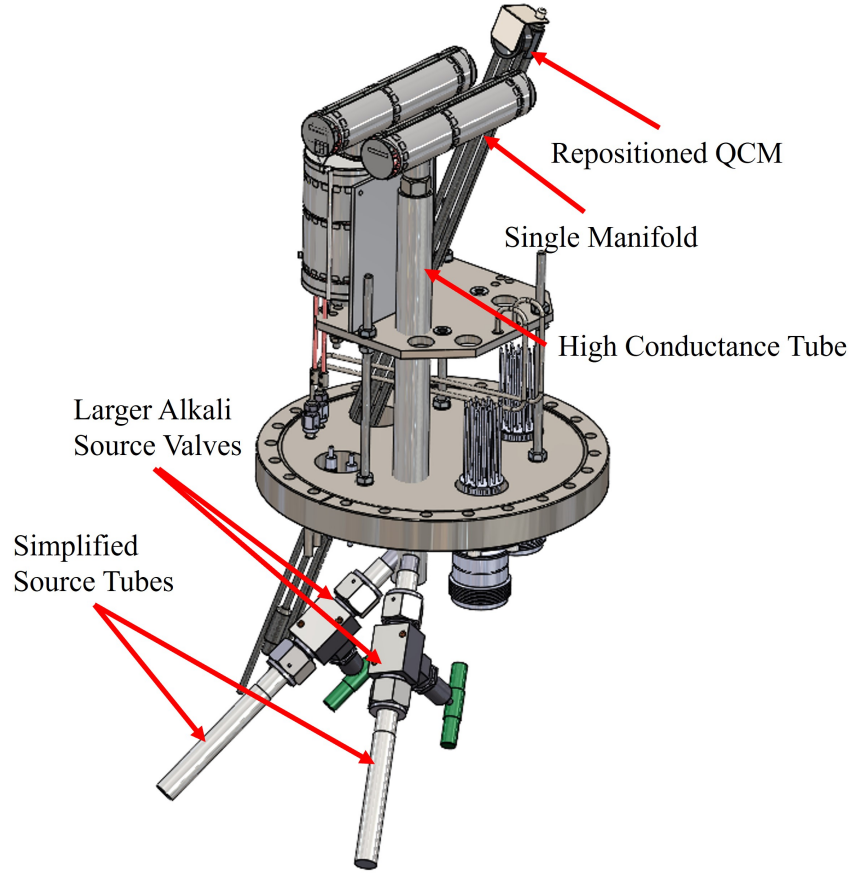


Figure 6.15: *A revised version of the source with a focus on limiting excess material inside the chamber, high source conductance, and improved alkali flux measurement. Much of the current source assembly could be repurposed for this design.*

is performed to ensure hermeticity prior to cathode growth. Copper pinch tubes, brazed to the detector housing, are used to slowly introduce alkali-metal vapor which diffuses over the cathode surface to form the sensitive film³². This process has many advantages over the current method — specifically, atmospheric assembly, the ability to perform leak testing prior to cathode growth, and a deposition volume limited to the detector itself. Employing these methods to the proposed reflective or transmissive PMT designs would enable higher yield and throughput. A batch production process, such as those used in commercial device manufacture (Fig. 6.16) could also be leveraged. Many of the vacuum and deposition components used in the current system could be reconfigured to support this process.

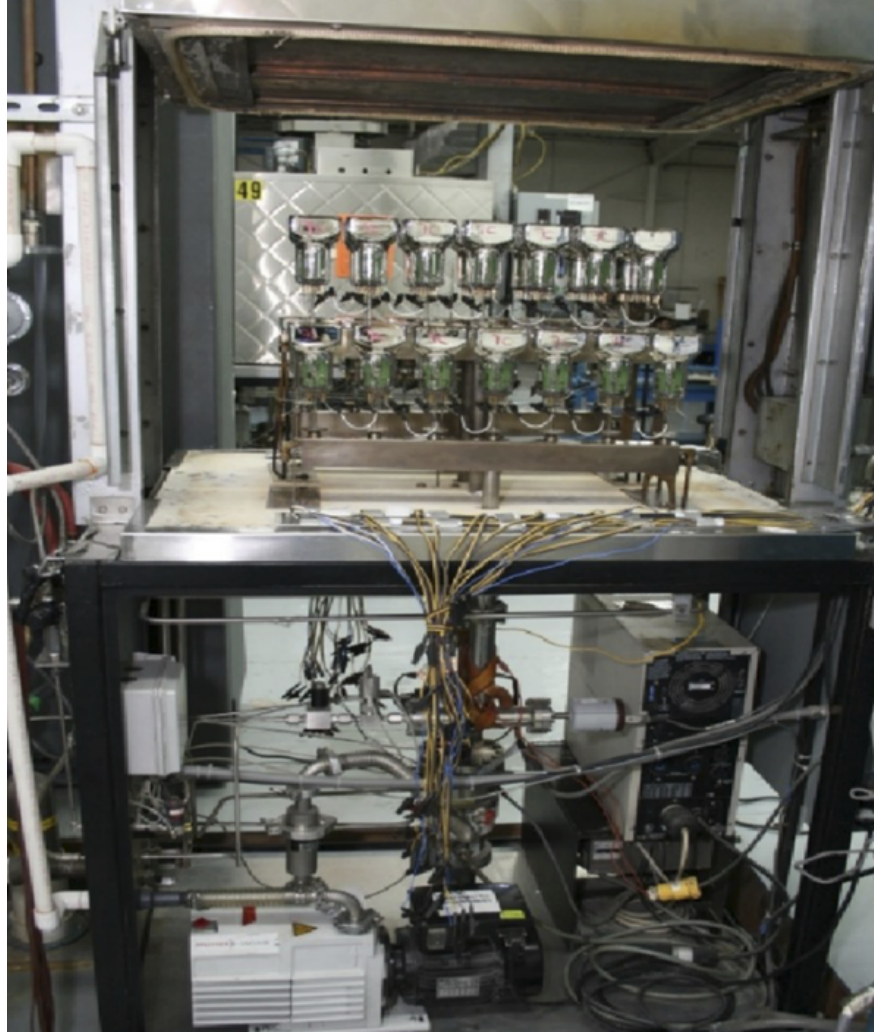


Figure 6.16: *A batch PMT production chamber formerly used by Photonis was sold to the LAPPD group for the development of their photocathode growth process³².*

6.4 Final Conclusions

The spatially-dependent response of photomultiplier tubes and scintillation spectrometers were measured to inform Monte Carlo studies of coupled gamma-optical photon transport for novel gamma-ray detectors. These measurements also served to elucidate the origins of spectrometer resolution and validate the standard, simplified models of detector performance. It was found that both the photodetector and scintillator responses were asymmetrically nonuniform, and optimization of the crystal and PMT orientations could provide lower energy resolution. For thin crystals, interactions occur in close proximity to the photocathode,

and photoelectron emission is somewhat localized to the gamma-ray absorption location. This means that the resolution is dependent upon both the scintillator and PMT responses near the interaction location. High aspect ratio crystals undoubtedly suffer from similar scintillation yield nonuniformity along their length; however, spatially dependent photon-transport characteristics may be tuned by modifying surface finish and reflector thickness based on the results of beam irradiation studies. Furthermore, photoabsorption and emission in high aspect ratio crystals is confined to a smaller photocathode area, improving photoemission and anode response uniformity. To support large frontal-area crystals needed for applications seeking high detection efficiency, a combination of uniform detector response and photon-transport optimization is required. A novel photomultiplier geometry utilizing a large rectangular optical interface has been proposed to improve photon collection and energy resolution; simultaneously leveraging the high photon yield of scintillator materials to reduce overall size and power consumption compared to standard large circular-window PMTs.

Empirically-based stochastic electron-emission models were developed and implemented in COMSOL Multiphysics using external subroutines which were called by the coupled electrostatic/charged particle tracing modules to define emission yields and energies for photo- and secondary electrons. Radiographic imaging techniques were used to reconstruct the geometry of a well-documented ET Enterprises 9214B PMT. The phenomenological simulations effectively modeled the nominal multi- and single-photoelectron responses of the commercial device and were subsequently implemented to enable iterative design and predictive performance analyses of two novel PMTs based on the rectangular-window geometry.

Based on the initial results of the design process, modeling and fabrication of componentry for a reflective photocathode, “Catoptric”, PMT were undertaken. Several stainless steel housings were machined from billet, and additional assembly of focusing and retaining structures within the vacuum envelope was successful. Further investigation of housings formed from pre-polished stainless steel or kovar are necessary to reduce batch production costs.

Metalization of 10 machined borosilicate optical windows and frames for electrical feed-

throughs was outsourced to commercial vendors. Dozens of glass frit bonds were attempted on test coupons and windows to characterize the bonding process. Early single-sided screen print attempts yielded few structurally-sound bonds due to insufficient wetting of the uncoated substrate. Additionally, attempts to bond windows and frames proved difficult due to the fragility of the frames. Double-coated assemblies provided a significant structural improvement over previous iterations. Glass wettability is greatly improved by coating both substrates with the frit paste, and the combination of powdered frit (post-burnout) in conjunction with bonding behavior between like-materials better adapted the frit to minor surface-nonuniformities over smaller boundaries. Still, microcrack formation in the electrical feedthrough, observed *post hoc*, preclude hermiticity. Additional 4 mm thick window surrogates were affordably fabricated using Kansas State University’s water jet cutter, however these samples also failed vacuum testing due to apparent frit-thickness nonuniformity. It is advised that an alternative frit with a lower melt-viscosity be sourced, or that the method be avoided entirely for large substrates in the future. Validated large-area indium sealing processes for photomultiplier applications have been identified, although few studies have been performed for the current work. Further investigation is needed.

Two methods of miniature-dynode structure fabrication, passivation, and metalization of electrodes were explored. The first attempts used wire-EDM aluminum structures with an oxide formed on the surface through anodization to enable electrically-isolated dynodes via subsequent E-beam deposition of Al/Ti/Au. The porous oxide is undesired for UHV applications and seemingly could not maintain discontinuity between electrodes after metalization. A second attempt sought to avoid the passivation step by additively manufacturing dynodes from a “non-outgassing”, high-temperature ceramic-like polymer. Subsequent E-beam evaporation and ENIG processes showed that the substrate softened significantly under heat, while additional research revealed substantial outgassing under vacuum. Should it be necessary, exploration of nonporous insulating coatings, such as those produced via ALD is advised.

A laboratory and UHV chamber to support sensitive material deposition and component assembly were constructed based on similar work from Argonne National Laboratory.

A deposition area containing three individual sources allowed for dual-zone thermal control/monitoring, photocurrent measurement, and sequential Sb, K, and Cs PVD. A second area within the same chamber provided assembly and thermo-compression indium sealing capabilities. A cooling block was incorporated to prevent photocathode degradation should seal-processing temperatures be too high. Initial photocurrent-measurement system characterization was successfully performed on a bulk silver substrate. Multiple photocathode depositions were attempted in an effort to parameterize the deposition process; however, source design, leakage current, and material/vacuum contamination hindered sensitive-material performance and characterization. Leakage current and material contamination are somewhat easily rectified by installing additional shielding and replacing the alkali source. Even so, the deposition source and vacuum chamber limitations may prohibit the growth of high-performance emissive films. A few modifications were proposed which may solve or reduce the impact of these issues.

However, given the complexity of several described fabrication processes, as well as the relatively poor photoelectron-collection capability and instability of the reflective design, it is recommended that a simpler, more robust transmissive PMT design and fabrication process be pursued. The additional QE obtained through reflective-photocathode growth may not outweigh the increased collection-efficiency of the transmissive design, especially considering Fresnel losses, dynode obstruction, and dimensional tolerance instability. By removing the dynode and focus structures from the optical path, far superior focus-electrode and dynode designs may be implemented. Standard all-metal electrodes may affordably be laser patterned from stainless steel, BeCu, or nickel sheet and formed in-house. Additional space, below the front-end structures, may be utilized to accommodate electrical connections and getter materials to maintain UHV for extended lifetime. The straightforward indium sealing process outlined by the LAPPD project may be adapted, enabling pre-cathode-deposition vacuum testing imperative to high detector throughput. Furthermore, the dual-vacuum assembly and deposition system is well described. The pre-deposited Sb/air-transfer method greatly simplifies the assembly process, while alkali-diffusion-based photocathode-growth results from Incom, Inc. reveal highly uniform film performance. Multiple sensitive-

material depositions may be attempted and reattempted within the same unit by diffusing the alkali constituents out of the thin film at elevated temperatures. The photodetector itself is the only chamber exposed to alkali materials, greatly simplifying cleanup and increasing throughput. Multipin electrical feedthroughs, purchased from commercial vendors, may be welded to the metal housing for prototype and/or batch production units, further simplifying construction. Much of the current vacuum system may be maintained for UHV Sb deposition, and extraneous components may be adapted for a second dual-zone vacuum assembly and alkali-deposition chamber.

6.5 Scientific Contributions

High Spatial Resolution Spectrometer Characterization The spatially-dependent anode response of an ET Enterprises 9490B PMT was measured using 545 nm light to inform coupled gamma-ray and optical photon transport simulations. The results of these studies are being used to quantify how PMT response nonuniformity affects scintillation spectrometer resolution for different crystal geometries. Similar studies have been performed using PMTs and cylindrical scintillators of several sizes^{73;131;132}; nevertheless, additional studies are needed to understand the consequences of photocathode nonuniformity and multiplier variance for high-efficiency rectangular crystals of various sizes and materials.

A collimated ^{137}Cs gamma-ray source was also constructed to provide validation data sets for several unique scintillator geometries and materials. The results presented in Ch. 2.2 are being used to tune optical transport parameters such as reflectivity and attenuation for coupled gamma-ray and optical photon transport simulations. Results for the "wide" $25.4 \times 127 \times 25.4 \text{ mm}^3$ CsI(Tl) crystal reveal a unique spatial dependence, meaning both the scintillator PMT response uniformity should be tailored to achieve fine energy resolution. PMT response is especially important for thin crystals and lower-energy gamma rays ($< 200 \text{ keV}$) which reduce R_i and have a tighter photon emission distribution⁷³. The "long" $25.4 \times 25.4 \times 127 \text{ mm}^3$ CsI(Tl) crystal measurements reveal localized light-transport nonuniformity caused by poor reflectance. This nonuniformity diminishes with distance from

the photocathode as the light distribution becomes more uniformly distributed. Combined, PMT and spectrometer nonuniformity studies provide a powerful tool for predicting novel spectrometer performance prior to fabrication.

Simulation of Photoemission in COMSOL The outlined method of sampling photoelectron emission energy improves on previous models which assign fixed energies of 0, 1, or 2 eV^{18;83;133}, as well as others which have sampled somewhat arbitrary distributions from 0-3 eV^{77;82}. Recent work by Achterman¹³⁴ implemented a similar photoelectron energy sampling method that was independently developed concurrently with the present work. Achterman also provides a comparison of 0 eV photoemission to varying momentum for several virtual monochromatic light sources ranging from 360-650 nm. The results clearly show that the spatial and time distributions of photoelectrons at the first dynode vary substantially for releases from the center of the photocathode due to low extraction field gradients at the cathode surface¹³⁴. Since this sampling method is wavelength-dependent, the sampled energy distribution is photon-spectrum agnostic, meaning that any light source may be used to develop a unique photoelectron energy distribution — provided the incident spectrum lies within the energies tested by Nathan and Mee²⁵.

Simulation of Secondary Electron Emission in COMSOL A novel method of sampling the three classifications of secondary electrons in COMSOL has also been implemented. Similarly to photoelectron emission, many PMT simulations simplify secondary electron emission yield and energy distributions due to limited data or difficulty incorporating these distributions into simulation software^{75;83;133;134}. This work uses external C subroutines called by COMSOL for each incident primary electron to sample the true secondary electron yield and energies. Accurate sampling of both SEY and energy are needed to fully model critical performance characteristics such as ENF and photoelectron collection efficiency.

Rectangular PMT Designs Two novel PMT concepts with large, rectangular optical windows and reduced gain were developed through iterative design using the aforementioned

simulation methods. The Catoptric PMT leverages a reflective photocathode to improve light distribution, QE, and response uniformity. A miniaturized dynode set limits photon absorption at the window, while maintaining an acceptable ENF, although the transfer and multiplier resolutions suffer due to reduced photoelectron collection efficiency. Simulations revealed relatively fast timing characteristics at an operating potential of 750 V.

A second transmissive photocathode PMT concept was developed to address many deficiencies associated with the performance and fabrication of the reflective design. The transmissive PMT design offers much better response uniformity across the photocathode owing to the large focus and dynode structures. High interstage collection efficiency increases multiplier gain with an ENF comparable to that of the Catoptric PMT for the same operating potential of 750 V. Timing characteristics are acceptable for fast scintillators, and further development of the focus and dynode structures will reduce the transit time spread induced by low extraction field gradients at the cathode periphery and dynode surfaces.

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Appendix A

Review of Optical Transport in Scintillators

A.1 Refractive Index

The optical characteristics of any material are, in part, described by the complex index of refraction

$$\mathbf{n} = n + ki \tag{A.1}$$

The real part of the refractive index n may be used to calculate the angle of refraction as well as the angle of total internal reflection θ_{crit} using Snell's Law

$$n_1 \sin \theta_1 = n_2 \sin \theta_2, \tag{A.2}$$

$$\theta_{crit} = \sin^{-1}(n_2/n_1) \tag{A.3}$$

A fraction of incident light is reflected at any mismatched optical boundary. This phenomena is known as Fresnel reflection and is dependent upon the polarization. Reflectance for parallel (p) and perpendicular (s) polarized light are given by the Fresnel equations, which are plotted

in Fig. A.1:

$$R_s = \left| \frac{n_1 \cos \theta_1 - n_2 \cos \theta_2}{n_1 \cos \theta_1 + n_2 \cos \theta_2} \right|^2 \quad (\text{A.4})$$

$$R_p = \left| \frac{n_1 \cos \theta_2 - n_2 \cos \theta_1}{n_1 \cos \theta_2 + n_2 \cos \theta_1} \right|^2. \quad (\text{A.5})$$

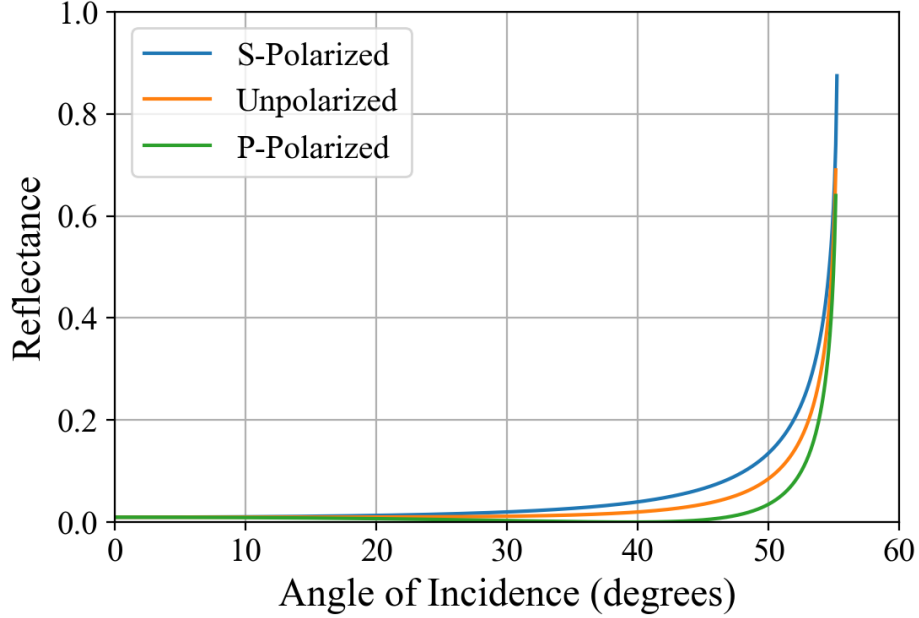


Figure A.1: Boundary reflectivity for $n_1 = 1.85$ and $n_2 = 1.52$, approximating a direct NaI(Tl)-glass interface. The critical angle is 55.2° .

A.2 Self-Attenuation

The attenuation of light through a medium follows the standard exponential attenuation $A = 1 - \exp(-\alpha * d)$, where A is fraction of light remaining at a distance, d , traveled through the material, and α is the absorption (or attenuation) coefficient. The absorption coefficient is related to the complex index of refraction by $\alpha = 4\pi k / \lambda_0$, where λ_0 is the vacuum wavelength. Alternatively, one may use absorption or attenuation length, λ_{att} (to avoid confusion with wavelength), which is the distance required to reduced intensity by $1/e$. Absorption length is simply the inverse of the absorption coefficient, $\lambda_{att} = \alpha^{-1}$. Absorption lengths from 1-5

eV for a few scintillators are given in Fig. A.2. For large-volume scintillators, where the path

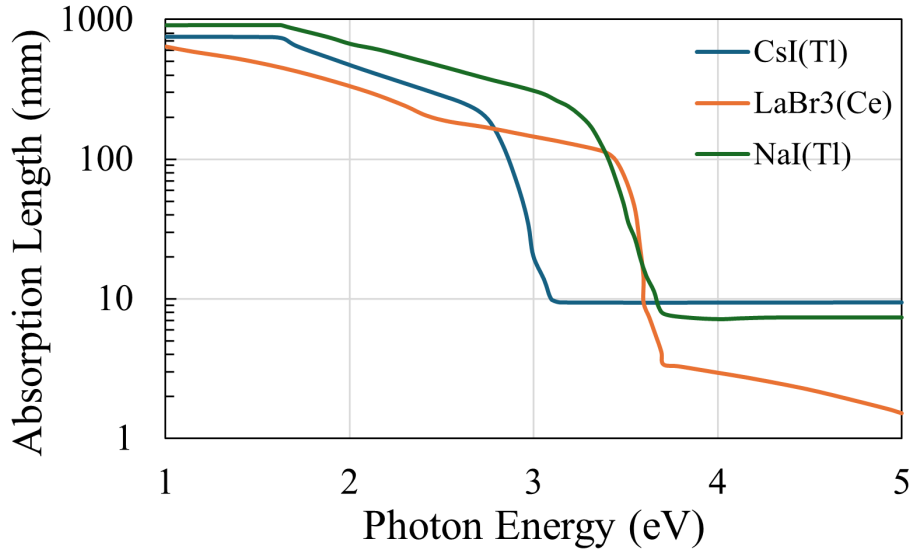


Figure A.2: The absorption lengths for CsI(Tl) , $\text{LaBr}_3(\text{Ce})$, and NaI(Tl) from Brown³³ (note errata).

a photon travels to reach the photocathode can be tortuous, the absorption length must be long over the emission energies such that photons are not absorbed by the scintillator, even after multiple reflections.

A.3 Reflectors and Optical Coupling

To enhance light collection, scintillators are surrounded by optical reflectors with various beneficial properties to promote light transport to the photodetector¹³⁵. The scintillator surface may also be textured in a way that promotes favorable photon transport^{136;137}. A mixture of specular and diffuse reflectors may be used to ensure consistent light collection independent of the interaction location. PTFE and TiO_2 are common diffuse reflectors, while foils and Enhanced Specular Reflectors (ESRs) are used as a backing reflector or directly applied to the crystal, when appropriate¹⁸. Commonly used reflectors in both categories will have a reflectivity $> 90\%$ over the emission wavelengths¹³⁵.

At the optical interface, a loss of 10-30% of the optical photons is standard due to suboptimal coupling, i.e. boundary reflections and light leakage¹⁸. Many scintillators are

hygroscopic, meaning they cannot be exposed to humid environments. This property enforces some limitations on packaging and optical coupling. Hygroscopic scintillators must be hermetically sealed in a dry environment. Demountable crystals are packaged separately from a photodetector, requiring an extra optical window besides that of the photodetector further degrading performance. Optical-coupling greases and epoxies with an intermediate index of refraction are employed to reduce photon reflections due to the refractive-index mismatch between a scintillator and glass. Couplants also reduce losses caused by imperfect surface finishes or gaps that would otherwise be filled with air. Dow Corning, GE and others make premium, purpose-built optical coupling greases, oils, and epoxies, however other silicon-based lubricants are much more affordable and easily procured. A practical resource on optical couplants is provided by Analog Services, Inc.¹³⁸.

Appendix B

Statistics of Electron Multiplication

The following summarizes Wright's¹⁸ statistical analysis of an electron multiplier which is based on the previous work of Zworykin and Shockley^{45;139}.

Discrete probabilities with binary outcomes are prevalent throughout radiation detection systems (e.g. the probability of detecting a photon). Consider the probability that observing a “success” for a single sample is p , while the probability of not observing a success, known as “failure”, is $1 - p$. The probability that a specific number of successes, n , are observed, while $N - n$ are unobserved is $p^n(1 - p)^{N-n}$, where N represents the total number of samples. The probability that any specific n will be observed is greater than $p^n(1 - p)^{N-n}$ by a factor of ${}^NC_n = \frac{N!}{n!(N-n)!}$, which is the number of combinations in which n may be achieved from N . Multiplying these terms produces the binomial distribution

$$P(n; N, p) = \frac{N!}{n!(N-n)!} p^n (1-p)^{N-n}. \quad (\text{B.1})$$

For a sufficiently large number of samples with a relatively low probability of success, we find the rate of success is $r = Np$. Substituting r/N for p , B.1 becomes

$$\begin{aligned} P(n; N, r) &= \frac{N!}{n!(N-n)!} \left(\frac{r}{N}\right)^n \left(1 - \frac{r}{N}\right)^{N-n} \\ P(n; r) &= \frac{r^n}{n!} e^{-r}, \end{aligned} \quad (\text{B.2})$$

which is the Poisson distribution. In the standard notation, μ is used in place of r , such that

$$P(n; \mu) = \frac{\mu^n}{n!} e^{-\mu}. \quad (\text{B.3})$$

For a finite set of N measurements, one cannot find the true mean μ , however the expectation value, $E[X]$, of the set, X , may be estimated by

$$\langle x \rangle = E[X] = \frac{1}{N} \sum_{i=1}^N x_i \approx \mu, \quad (\text{B.4})$$

where x represents individual outcomes. The spread of a distribution may be described using the the variance. The variance is calculated using

$$\text{var}(X) = \frac{1}{N} \sum_{i=1}^N (x_i - \mu)^2 = x_i^2 - \mu_i^2, \quad (\text{B.5})$$

which for the Poisson distribution $\text{var}(X) = \mu$.

To more easily manipulate a discrete distribution — also known as a probability mass function (PMF) — we may use a power series representation of a PMF known as the probability generating function. The generating function $G(u)$ for any PMF $p(x)$, finite or infinite, is defined as

$$G(u) = \sum_{x=0}^n p(x) u^x. \quad (\text{B.6})$$

The probability distribution must be normalized to one, thus $G(1) = 1$. Differentiating [B.6](#) with respect to u provides

$$G'(u) = \sum_{x=0}^{\infty} x p(x) u^{x-1}, \quad (\text{B.7})$$

$$G'(1) = \sum_{x=0}^{\infty} x p(x) = E[X]. \quad (\text{B.8})$$

$G'(1)$ is the mean or expectation value of the set $E[X]$. Differentiating $G'(u)$ yields

$$G''(u) = \sum_{x=0}^{\infty} x(x-1)p(x)u^{x-2}; \quad (\text{B.9})$$

$$G''(1) = \sum_{x=0}^{\infty} x(x-1)p(x) = E[X(X-1)]. \quad (\text{B.10})$$

Using the resultant derivatives and recalling Eq. B.5, we may calculate the variance

$$\begin{aligned} \text{var}(X) &= E[X^2] - (E[X])^2 \\ &= E[X^2] - E[X] + E[X] - (E[X])^2 \\ &= G''(1) + G'(1) - (G'(1))^2. \end{aligned} \quad (\text{B.11})$$

For an electron multiplier, cascade theory may be used to calculate the signal dispersion brought about by sequential processes, where the electron multiplication of a previous stage affects the total output of the next. For a simple two-stage cascade from $A \rightarrow B$, the generating function is

$$G_{AB}(u) = G_A[G_B(u)], \quad (\text{B.12})$$

$$G'_{AB}(u) = G'_A[G_B(u)] \times G'_B(u), \quad (\text{B.13})$$

$$G''_{AB}(u) = G''_A[G_B(u)] \times G'_B(u)^2 + G'_A[G_B(u)] \times G''_B(u). \quad (\text{B.14})$$

Recall from B.6, B.8, and B.10 that generating functions have the following properties

$$G_i(1) = 1$$

$$G'_i(1) = E[X]$$

$$G''_i(1) = E[X(X-1)]$$

Therefore,

$$G'_{AB}(1) = G'_A[G'_B(1)] \times G'_B(1)$$

$$m_{AB} = m_A \times m_B, \quad (\text{B.15})$$

and

$$G''_{AB}(1) = G'_A[G'_B(1)] \times (G'_B(1))^2 + G'_A[G'_B(1)] G''_B(1)$$

$$\text{var}_{AB} = G''_{AB}(1) + G'_{AB}(1) - (G'_{AB}(1))^2$$

$$= \text{var}_A \times m_B^2 + \text{var}_B \times m_A. \quad (\text{B.16})$$

It easy to see from [B.15](#) and [B.16](#) that the mean and variance may be modeled for cascaded processes, such as signal dispersion as a result of multiplication through a dynode chain. The expected gain $\langle g \rangle$ after the k^{th} dynode stage is simply the product of the yield δ at each antecedent stage

$$\langle g \rangle = \prod_{i=1}^k \delta_i. \quad (\text{B.17})$$

The variance in gain, σ_k^2 , after stage k may be calculated with the mean yield δ_i and the variance of individual stages σ_i^2 using

$$\sigma_k^2 = \langle g \rangle^2 \left(\frac{\sigma_1^2}{\delta_1^2} + \frac{\sigma_2^2}{\delta_1 \delta_2^2} + \frac{\sigma_3^2}{\delta_1 \delta_2 \delta_3^2} + \dots + \frac{\sigma_k^2}{\delta_1 \delta_2 \delta_3 \delta_{k-1} \delta_k^2} \right). \quad (\text{B.18})$$

One can see that the largest contributor to signal variance is the first stage, as the influence of variance in yield for the latter stages is diminished by the increased number of electrons incident upon that stage. Assuming an equivalent mean yield δ and a Poissonian yield distribution^{[18;21](#)} for k multiplication stages, the mean gain is $\langle g \rangle = \delta^k$, and the variance at any individual stage is $\sigma^2 = \delta$. Because of this, calculating the total variance may be

simplified:

$$\begin{aligned}
\sigma_k^2 &= \delta^{2k} \left(\frac{1}{\delta} + \frac{1}{\delta^2} + \frac{1}{\delta^3} + \dots + \frac{1}{\delta^k} \right) \\
&= \frac{\delta^{2k}}{\delta} \left(1 + \frac{1}{\delta} + \frac{1}{\delta^2} + \dots + \frac{1}{\delta^{k-1}} \right) \\
&= \frac{\delta^{2k}}{\delta} \frac{\delta}{\delta - 1} = \delta^{2k} \frac{1}{\delta - 1} = \langle g \rangle^2 \frac{1}{\delta - 1}.
\end{aligned} \tag{B.19}$$

Typically, the photocathode-to-dynode voltage is higher than the interdynode voltage. This is done to improve electron focus, timing, and signal variance characteristics. Pulling out the first-stage yield δ_1 , the variance becomes

$$\begin{aligned}
\sigma_k^2 &= \frac{(\delta_1 \delta^{k-1})^2}{\delta_1} \left(1 + \frac{1}{\delta} + \frac{1}{\delta^2} + \dots + \frac{1}{\delta^{k-1}} \right) \\
&= \frac{(\delta_1 \delta^{k-1})^2}{\delta_1} \frac{\delta}{(\delta - 1)} = \langle g \rangle^2 \frac{\delta}{\delta_1 (\delta - 1)}.
\end{aligned} \tag{B.20}$$

Thus, the fractional variance is

$$\frac{\sigma_k^2}{\langle g \rangle^2} = \frac{\delta}{\delta_1 (\delta - 1)}. \tag{B.21}$$

Appendix C

9490B PMT Active Voltage Divider Circuit Diagram

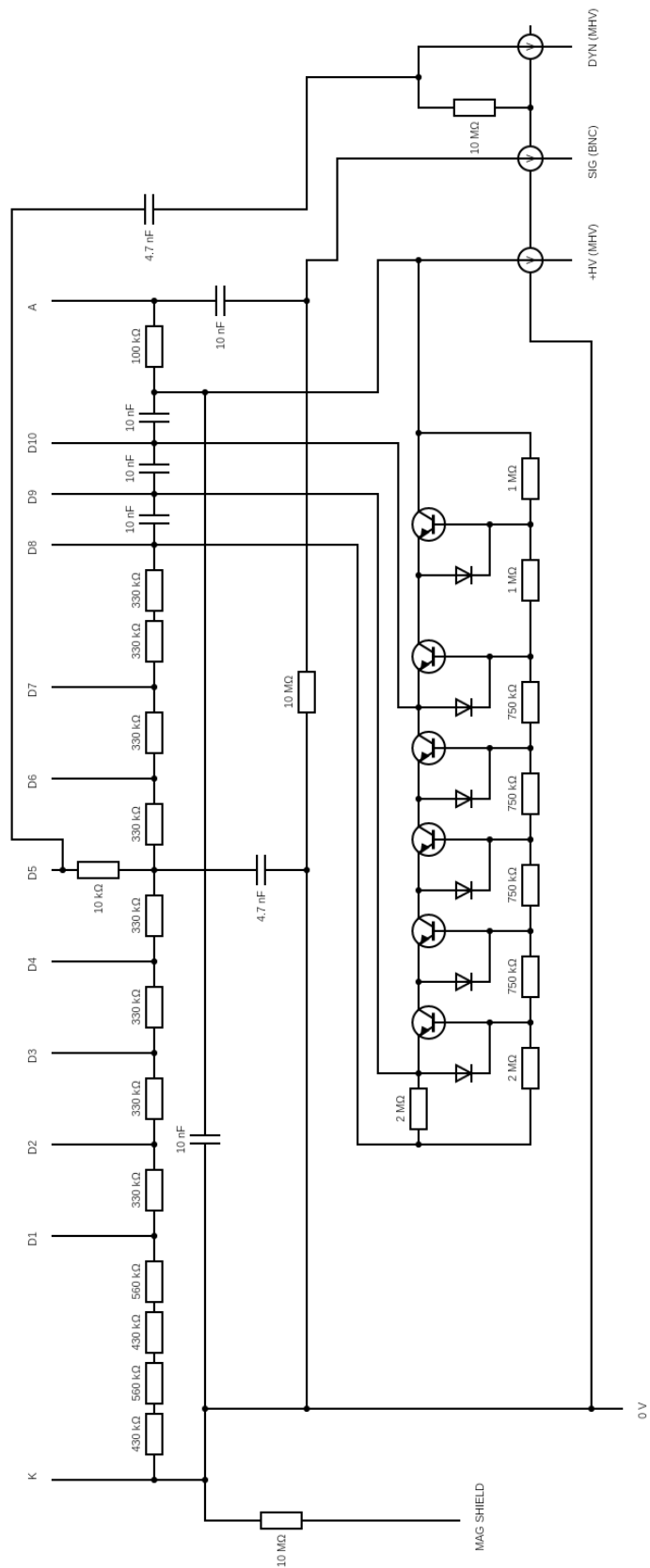


Figure C.1: Circuit diagram for the custom ET Enterprises 9490B voltage divider with D_5 readout.