

Development of an Instrument for Spatially Resolved, Optically Stimulated
Luminescence Dosimetry of Cobble and Dosimeter Surfaces

by

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July, 2023

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Optically stimulated luminescence (OSL) dosimetry is a method used to determine the amount of energy stored within a crystalline insulator due to ionizing radiation. At its most fundamental, OSL dosimetry requires optical stimulation to induce a sample to emit luminescence, a light detection apparatus to collect the luminescence signal, and a calibrating radiation source to convert the acquired signal into an equivalent dose. Conventional instruments have successfully integrated these components to perform OSL dosimetry on sediment and dosimeters. In this dissertation, an instrument was developed that improves upon commercially available instruments by providing spatial information that allows dose-mapping. The novel instrument LuCIDD (Luminescence instrument with Confocal and Imaging unit for Dating and Dosimetry) is based on the principles of a confocal microscope. This dissertation outlines the requirements for spatially resolved dosimetry mapping and describes the design and construction of LuCIDD. Tests of LuCIDD's lasers' ability to perform spatially resolved stimulating measurements were made by measuring their focal spot size, power density, and penetration depth. Used for calibration, the built-in X-ray source's energy spectrum, uniformity, and dose rate were characterized. The minimum resolution and stimulation time of measurements were determined, quantifying the amount of time to complete a dose map of a sample's surface. Lastly, LuCIDD's ability to recover a known applied dose from single points was verified to provide a proof-of-concept for future dose-mapping measurements.

Development of an Instrument for Spatially Resolved, Optically Stimulated
Luminescence Dosimetry of Cobble and Dosimeter Surfaces

A Dissertation

Presented to the Faculty of the Department of Physics
East Carolina University

In Partial Fulfillment of the Requirements for the Degree
Doctor of Philosophy
in Biomedical Physics

by

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July, 2023

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Acknowledgements

I must extend my deepest gratitude to my research advisor Dr. Regina DeWitt for the support, guidance, and constructive criticism throughout this research process; without her, my success would not have been possible. I'm also extremely grateful to my committee members who have contributed valuable insight into my research, as well as my career.

Special thanks to Chris Bonnerup, Gene Oakley, William Holland, and Dr. Ken Jacobs who have assisted with the software development, machining, and electronics troubleshooting for this research.

Finally, I could not have undertaken this journey without the support of my delightful wife who drove me to stay the course when times were rough, and shared the joy when times were good.

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Chapter 1

Introduction

1.1 History of Luminescence Dosimetry

Luminescence is the process of stimulating the emission of photons from a crystal which has stored external energy, usually in the form of a radiation dose. Luminescence finds its roots with Sir Robert Boyle who boasts the first recorded account of the phenomenon in 1664 when he held a borrowed diamond “a good while upon a warm part of my Naked Body” until this “brought it to some kind of Glimmering Light” (Boyle 1664). This description came two centuries before the term luminescence was even coined. The lack of early progress in this field is due to the quantized nature of luminescence, leaving the mechanisms that govern it unclear until the early twentieth century. It wasn’t until this time that quantum mechanics uncovered the fundamental properties of electrons, allowing descriptions of electron “movements” in materials – particularly crystalline insulators. Since then, the field of luminescence dosimetry has grown wildly with applications that range from medical to space and has been responsible for the development of an entire branch of archaeological and geological dating (Huntley, Godfrey-Smith, and Thewalt 1985; Aitken 1985; Aitken 1998; Yukihiro and McKeever 2011; Rhodes 2012).

Optically stimulated luminescence (OSL) dating makes use of visible light to cause

samples to emit luminescence and is commonly used on sediments to obtain information on a range of geological and archaeological topics. Ages up to several hundred thousand years old can be found when dating quartz and feldspar in sedimentary deposits. These sediments tend to be abundant in nature and have the best properties for OSL dating: a bright OSL signal that is easily reset and trapped electrons that remain stable over long periods of time. In 2000, Murray and Wintle published a standardized methodology for dating sediments using OSL and further refined their dating procedure with an article in 2003. Using a combination of laboratory irradiation, heating, stimulation, and measurement, Murray and Wintle were able to calibrate the natural signal of samples against a laboratory radiation source to develop dose response curves. This dose response curve allowed them to determine the “equivalent dose” of the sample - the amount of laboratory irradiation required to cause the sample to emit the same amount of luminescence as the natural signal. Finally, dividing the equivalent dose by the sample’s natural environmental dose rate allowed the calculation of the time since the sample had last been “bleached,” or when it had last emptied its stored energy due to sunlight exposure.

As luminescence dating of sediments saw widespread use around the world, researchers began to venture into areas lacking suitable sediments. The solution to this new problem was surprisingly intuitive: if luminescence dating works on sediments, then one should be able to apply it to rock surfaces. Liritzis (1994) was the first to suggest the use of thermally stimulated luminescence (TL) to date ancient stone structures, and the process was applied to the Apollo Temple at Delphi as well as two pyramids in Greece (Liritzis et al. 1997; Theocaris, Liritzis, and Galloway 1997). Unfortunately, residual TL signals were observed even after the sample was exposed to sunlight, causing large uncertainties in the calculated age of the structures. This led to investigations on the penetration of sunlight into surface materials to determine if TL dating was viable for rock surface dating (Liritzis and Galloway 1999; Simms et al. 2011; Fuhrmann et al. 2022). OSL was found to circumvent this issue entirely, acquiring almost no residual OSL signal

at meaningful sample depths (Habermann et al. 2000).

1.2 Dissertation Motivation

As cobble surface dating was investigated in more depth, there was a want for spatially-resolved data which would allow researchers to retrieve information from surfaces without separating the minerals and destroying the spatial data within. This resulted in advancements in instrumentation that were qualified to produce images with two-dimensional resolution (Habermann et al. 2000; Bailiff and Mikhailik 2003; Greilich, Glasmacher, and Wagner 2002; Greilich and Wagner 2006). Using lasers or LEDs to stimulate luminescence and CCD chips or photomultiplier tubes to collect signal, these instruments are able to make measurements of cobble surfaces with high spatial resolution (250 μ m - 25 μ m).

These instruments may be able to obtain spatially resolved data, but they are unable to separate the surface signal from the signal originating below the surface of the cobble, requiring samples to have been bleached to depths of up to 4 mm prior to deposition (Habermann et al. 2000). The current 2D instruments also lack multiple components essential to the luminescence and dating process: they contain no heat nor radiation source and are only able to stimulate and record luminescence. Because of this, samples need to be repeatedly transported between the measuring facility and the heating and irradiation facilities. This makes it difficult to replace the sample in the measuring facility in the exact same position, leading to spatial errors. With cobble surface dating finding more applications, the need for a more robust measurement instrument is clear.

The aim of this dissertation is to develop an instrument that increases the robustness of OSL dosimetry of cobble surfaces in four ways: 1) by allowing the heating, irradiation, stimulation, and measurement of a sample without removing it from the instrument, 2) by increasing the number of stimulation sources to allow for measurement of more varied samples, 3) by increasing the resolution of measurements in all three spatial dimensions,

and 4) by replacing the conventional beta radiation source with a more uniform and easier to store portable X-Ray tube. This was accomplished through a novel instrument for luminescence dosimetry named LuCIDD (Luminescence instrument with Confocal and Imaging unit for Dating and Dosimetry). The goal for LuCIDD is to allow the measurement of spatially resolved dose information from surfaces with non-uniform thickness. These “dose maps” of surfaces can be used for applications in space and medical dosimetry as well as cobble dosimetry and dating. LuCIDD’s components were characterized to determine their effectiveness for performing luminescence measurements, and LuCIDD was tested to provide a proof-of-concept for high resolution dose mapping measurements.

1.3 Dissertation Outline

This section outlines the following chapters in this dissertation. **Chapter 2** describes the physical basis of crystalline energy bands that allow the phenomenon of luminescence to exist. **Chapter 3** discusses applying luminescence to OSL dosimetry and dating using conventional instruments and measurement techniques. Challenges of applying conventional OSL dosimetry methods to cobbles are discussed in **Chapter 4**, along with changes in instrumentation used to circumvent these challenges. **Chapter 5** outlines the construction of the novel luminescence instrument LuCIDD that has been built, characterized, and tested during the course of this dissertation to provide spatially resolved equivalent dose information on sample surfaces.

Chapter 6 tests and characterizes the focal spot size, power density, and penetration depth of LuCIDD’s stimulating lasers to determine their applicability to performing spatially resolved measurements. LuCIDD’s X-ray was characterized for energy, uniformity, and dose rate in **Chapter 7**. In **Chapter 8**, the optimal stimulation time and minimum resolution for ideal measurements was determined, and LuCIDD’s ability to measure a known applied dose was verified. Finally, **Chapter 9** provides a summary and discussion

of these results, as well as multiple suggestions for future research directions.

Chapter 2

Physical Basis of Luminescence

Dosimetry

Optically stimulated luminescence (OSL) is a direct product of the energy band structure that can be found in crystalline solids. Incoming ionizing radiation causes energized electronic movement throughout the energy bands, giving electrons the opportunity to store their increased energy within the crystal. When an energized crystal is exposed to stimulating light, the stored energy is released as photons – the OSL signal. This signal is proportional to the absorbed ionizing radiation, allowing recorded OSL to be used for dosimetric purposes.

Before details of LuCIDD are discussed, it is necessary to explain the underlying physics of the luminescence process. This chapter lays the foundation required to understand the origin of the OSL signal by detailing the energy band structure of crystals and exploring the mathematical basis of OSL.

2.1 Structure of Crystalline Solids

In 1913, W. H. Bragg and W. L. Bragg became the first to experimentally verify the periodicity of crystal lattices using X-ray crystallography. These crystal lattices may be expressed in terms of a “primitive cell” that repeats periodically throughout the solid. The periodicity of the primitive cell generates the energy band structure that governs the phenomena of luminescence. This section describes how energy bands arise from the periodic primitive cell.

2.1.1 Energy Bands in Crystals

Electrons that are bound to atoms are forced to have discrete energies called “levels” in which only a single electron may occupy. These singular energy levels are split and multiplied when multiple atoms are arrayed into a crystal lattice. If enough atoms are combined, the resulting energy levels are so closely spaced they form quasi-continuous “bands” of allowable energies, separated by “gaps” of forbidden energies (Fig. 2.1). Energy levels, and thus energy bands, arise from the interactions between electrons and the nuclei they are bound to. For the simplest three-dimensional case, the hydrogen atom, the potential $V(\mathbf{r})$ some distance $|\mathbf{r}|$ away from a single proton is:

$$V(\mathbf{r}) = -\frac{1}{4\pi\epsilon_0} \frac{q}{|\mathbf{r}|} \quad (2.1)$$

Here, ϵ_0 is the permittivity of free space and q is the charge of the particle. To find the possible energy levels, one must look to Schrödinger’s time-independent wave equation:

$$\left[-\frac{\hbar^2}{2m} \nabla^2 + V(\mathbf{r}) \right] \Psi(\mathbf{r}) = E\Psi(\mathbf{r}) \quad (2.2)$$

Here, $\hbar$ is the reduced Planck’s constant, m is the mass of the electron, ∇^2 is the Laplacian, $\Psi(\mathbf{r})$ is the electron’s wave function, and E is the total energy of the system. With

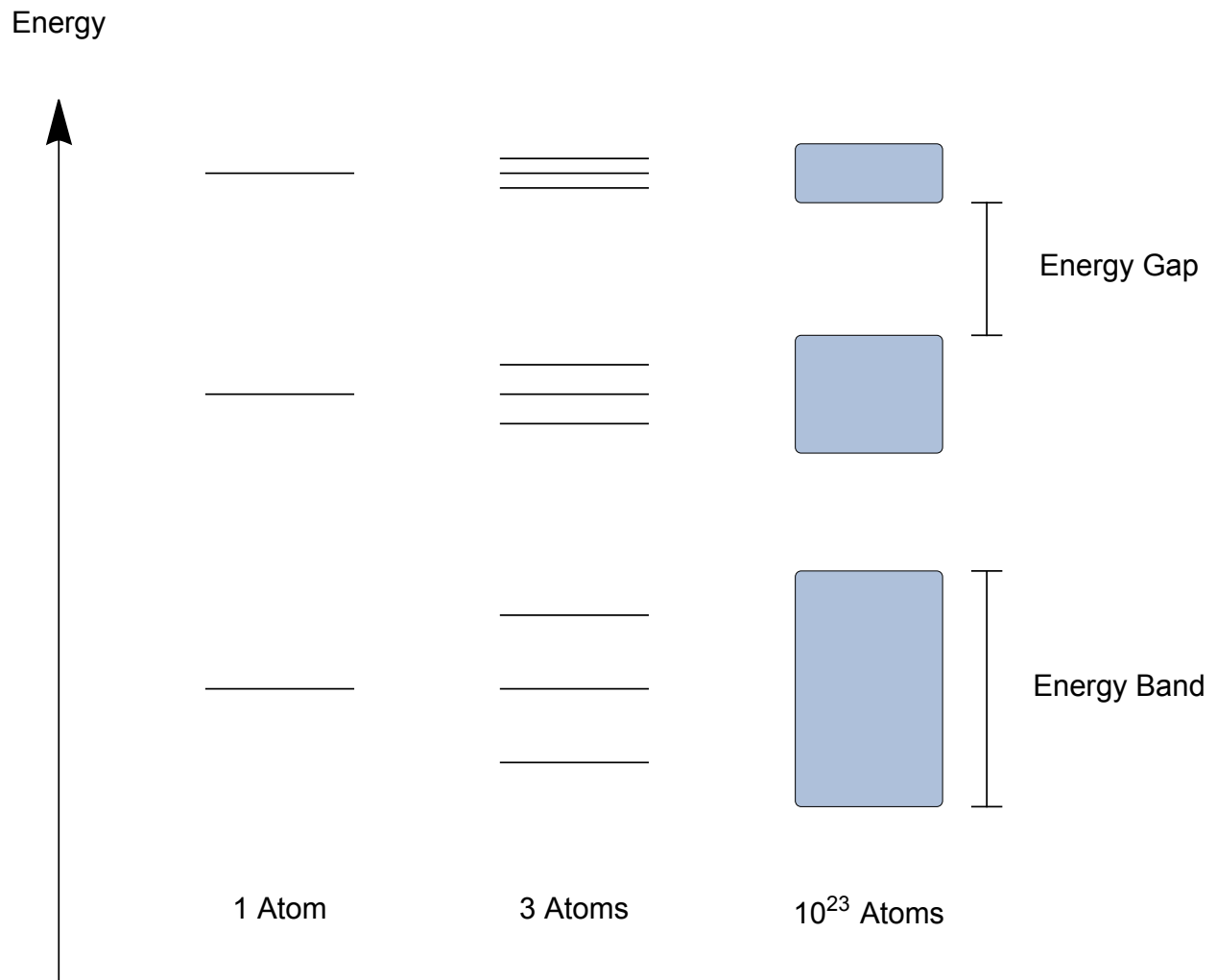


Figure 2.1: Illustration of energy levels, bands, and gaps in a crystal lattice. As more atoms are combined into a lattice, more energy levels are created until they form quasi-continuous bands of energies. The horizontal lines represent discrete energy levels, while the blue boxes represent energy bands.

great labor, the energy eigenvalues of the three-dimensional hydrogen atom are found to be:

$$E_n = -\frac{me^4}{8h^2\epsilon_0^2n^2} = -\frac{13.6}{n^2}\text{eV} \quad (2.3)$$

with e as the charge of the electron/proton, h as Planck's constant, ϵ_0 as the permittivity in free space, and n as the principal quantum number. Combining multiple atoms together into a periodic lattice generates a periodic potential which leads to Felix Bloch's 1929 solution to Schrödinger's equation, "Bloch's Theorem:"

$$\Psi(\mathbf{r}) = V(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}} \quad (2.4)$$

Where $\mathbf{k}$ is the wave vector, $e^{i\mathbf{k}\cdot\mathbf{r}}$ is the equation for a plane wave, and the potential is periodic over the primitive cell of the lattice with length $|\mathbf{a}|$:

$$V(\mathbf{r}) = V(\mathbf{r} + \mathbf{a}) \quad (2.5)$$

Kronig and Penney developed a model in 1931 to solve the simplest case - an infinite, one-dimensional periodic lattice with a potential of the form of equation 2.5. Their model devised a simplified square wave potential with interatomic spacing – and thus, potential well distance – a , potential well width b , and potential well depth V_0 (Fig. 2.2).

Because of its periodicity, solutions can be obtained for the regions inside ($-b < x < 0$) and outside the potential well ($0 < x < a - b$):

$$\cos(ka) = \cos(a\alpha) + P \frac{\sin(a\alpha)}{a\alpha} \quad (2.6)$$

Here, $\alpha = 2mE/\hbar^2$, $P = mV_0ba/\hbar^2$, and as $b \rightarrow 0$, $V_0 \rightarrow \infty$ while V_0b remains constant. The left-hand side of equation 2.6 is a cosine function that only covers a range of values from -1 to $+1$, but the right-hand side may exceed these values - thus there are values of

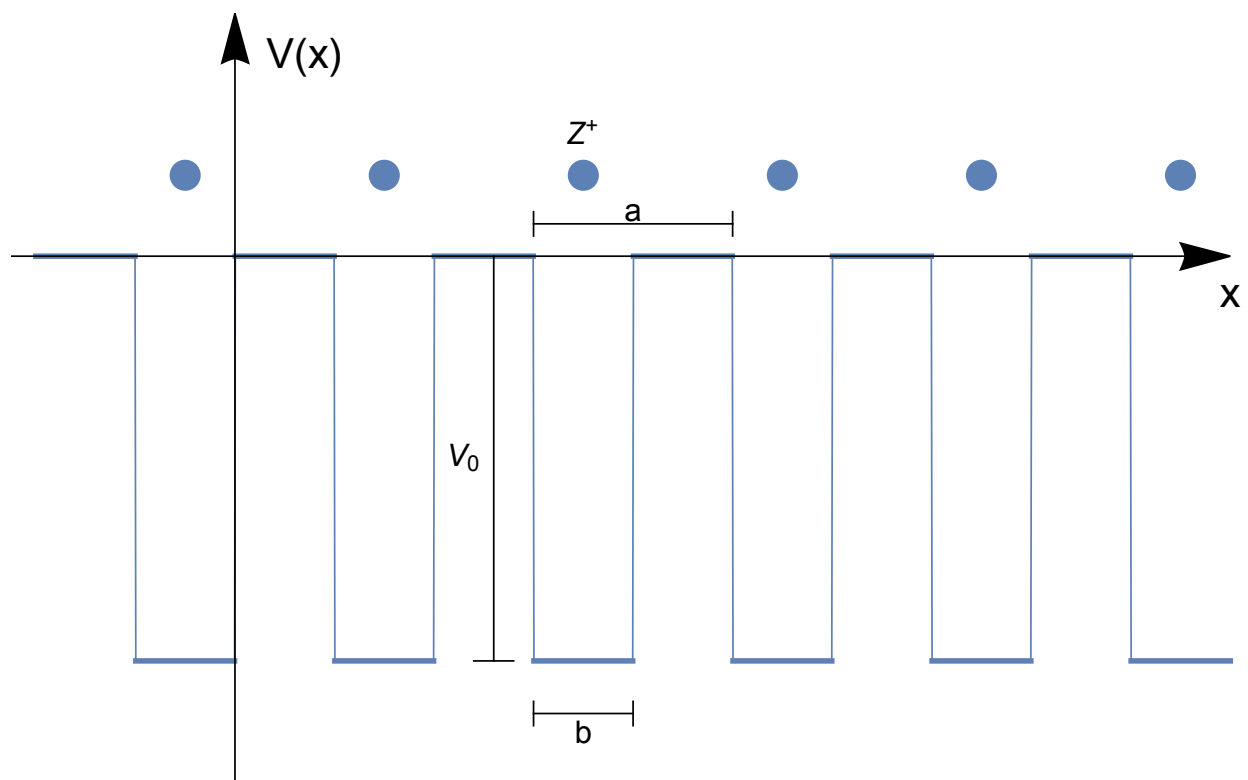


Figure 2.2: Illustration of a one-dimensional periodic square wave potential. Each potential well is caused by a proton in a periodic lattice.

$a\alpha$ that are not allowed, giving a null solution to Schrödinger's equation. These null solution values give rise to the forbidden energies i.e., the energy gaps of the band structure, while the allowable values of $a\alpha$ define the allowed energies in the energy bands.

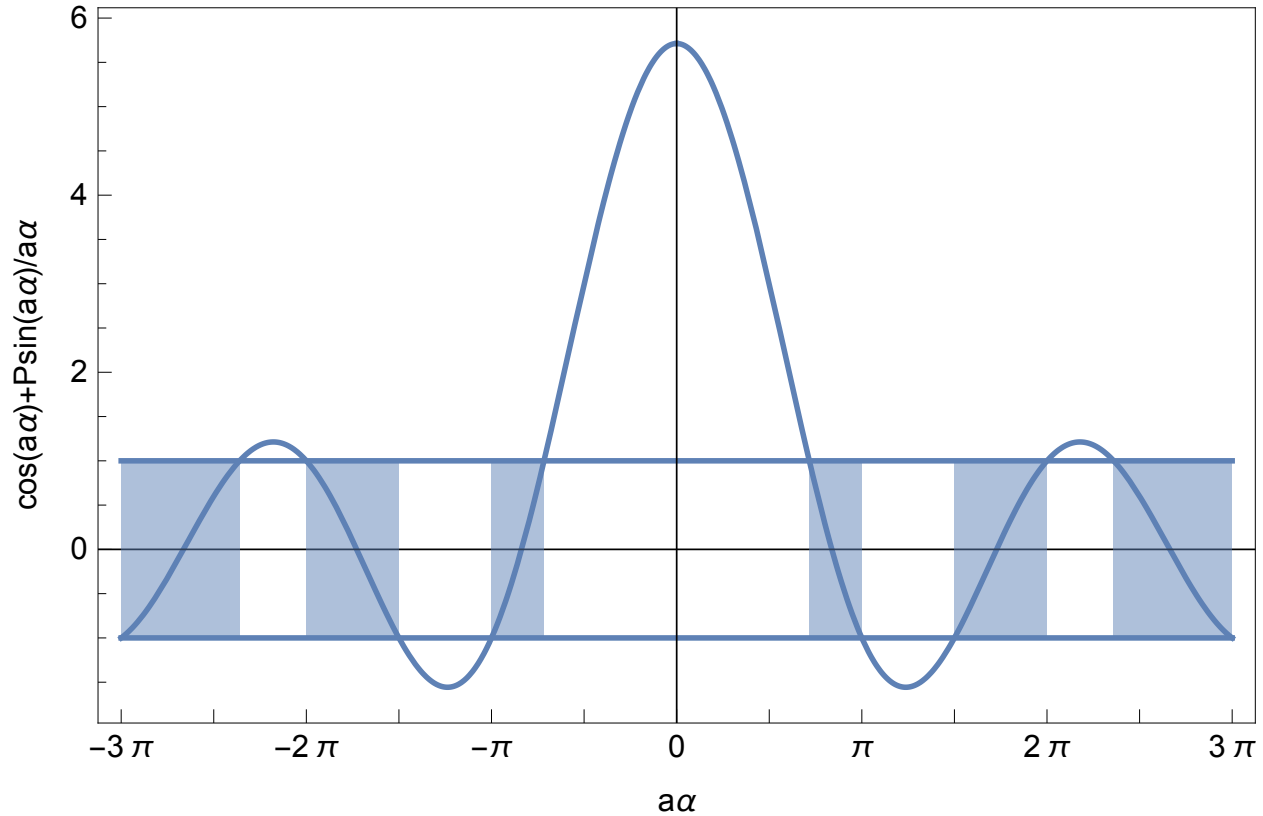


Figure 2.3: Plot of the Kronig and Penney model. The right-hand side of equation 2.6 is plotted against $a\alpha$ to display how equation 2.6 predicts the energy band structure of solids. The horizontal lines correspond to the upper and lower limits of the left-hand side of equation 2.6 and the shaded regions are where the values of $a\alpha$ are allowed as a solution to equation 2.6.

Figure 2.3 is a plot of the Kronig Penney model where allowable values of $a\alpha$ occur only within the shaded regions of the plot. These regions represent bands of allowable energies, while the unshaded portions of the plot produce energy gaps. As $a\alpha$ increases, the width of the energy bands increases, and the width of the energy gaps decreases.

The Fermi-Dirac distribution allows us to define attributes of a crystalline solid using its energy band structure:

$$f(E) = \frac{1}{e^{(E-\mu)/kT} + 1} \quad (2.7)$$

$f(E)$ is the probability that an electron will occupy an energy level with energy E . μ is the electro-chemical potential, k is the Boltzmann constant, and T is the absolute temperature. This probability distribution leads to the definition of the Fermi level - the energy E which provides a probability of occupation $f(E) = 1/2$ at thermodynamic equilibrium. The *filled* energy band closest to the Fermi level is called the valence band, while the nearest *unfilled* band is defined as the conduction band (Fig. 2.4). It is the size of the energy gap between the valence and conduction bands that defines the electrical properties of a crystal. Materials with no band gap are considered conductors and are exceptional at carrying electrical current. Materials with a band gap of <3.5 eV are called semiconductors. These materials are neither good insulators nor conductors, but their conductive properties are easily manipulated, lending to their use in high quality electronics. Lastly, materials with a band gap of >3.5 eV are called insulators, easily resisting electric current.

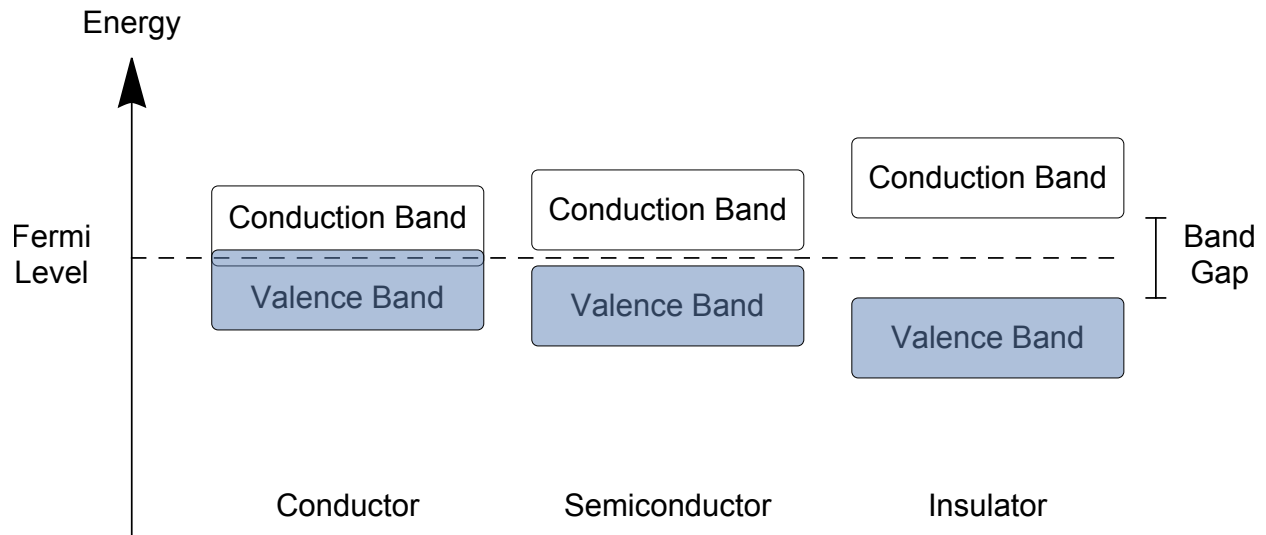


Figure 2.4: Illustration of the valence band, conduction band, band gap, and Fermi level of conductors, semiconductors, and insulators at $T = 0$. Shaded regions correspond to occupied electron states while unshaded regions correspond to empty states. For all solids, the Fermi level at $T = 0$ lies in the center of the band gap and the size of the band gap defines the solid as a conductor, semiconductor, or insulator.

Only perfect crystals have been discussed so far, and natural crystals rarely develop perfectly and without defects. These defects occur as a deformity of the lattice structure, missing ions, extra ions, or even foreign ions. Each of these defects adds a small, localized potential such that equation 2.5 gains a second term: $V(\mathbf{r}) \rightarrow V(\mathbf{r}) + V'(\mathbf{r})$ where $V'(\mathbf{r})$ is zero far from the defect. This leads to energy levels *inside* of the band gap which act as “traps” for mobile charge carriers (Fig. 2.5a). Mobile charge carriers occur when electrons are ionized from their atoms in the valence band into the conduction band. Upon ionization, electrons leave behind a hole - a net positive charge characterized by the *lack* of an electron (Fig. 2.5b) (Ibach and Lüth 2009). While not true particles, holes will freely travel through the valence band just as electrons travel freely through the conduction band. It is the ionization, trapping, and de-trapping of these charge carriers that causes the phenomenon of luminescence.

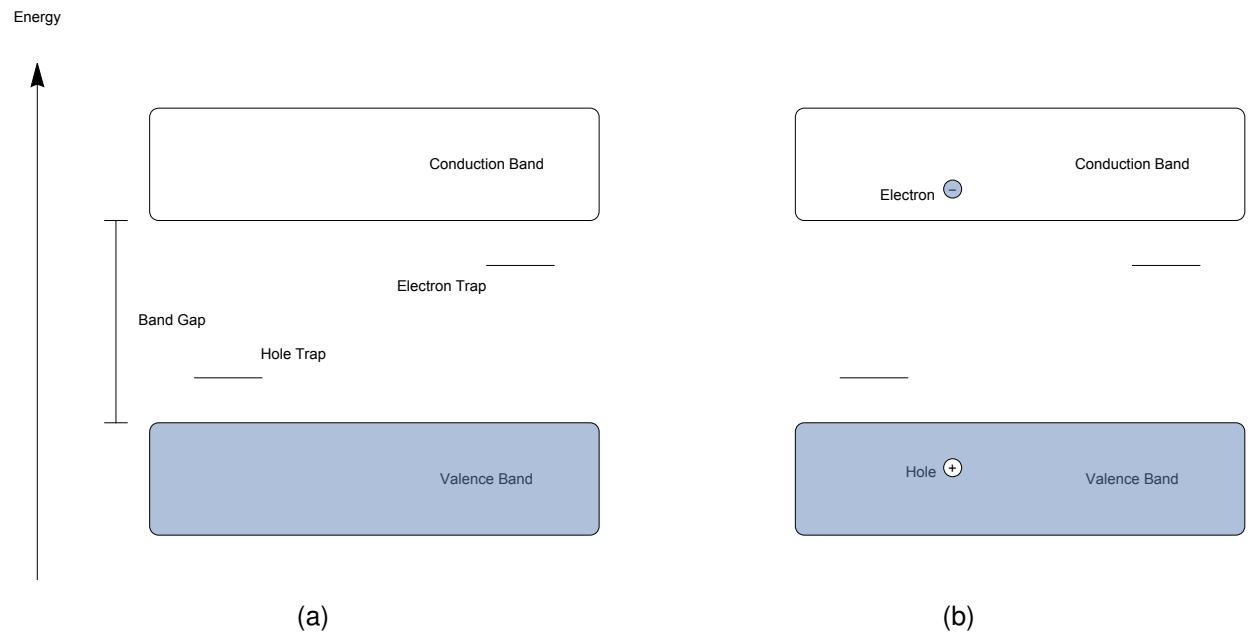


Figure 2.5: (a) Illustration of the electron and hole traps produced by defects in a natural crystal. (b) Illustration of an excited electron leaving a hole behind in the valence band. Both the excited electron and the hole are free to travel within their respective energy bands.

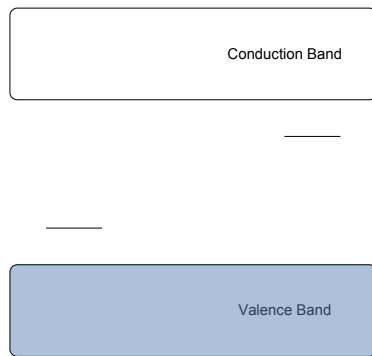
2.2 Luminescence Phenomena

Broadly discussed in the introduction, luminescence is the emission of light from a crystal that has stored external energy. Different types of luminescence are named for the way in which they are stimulated. For example, thermoluminescence (TL) applies heat to a sample to stimulate the emission of luminescence, while the subject of this dissertation - optically stimulated luminescence (OSL) - uses photons.

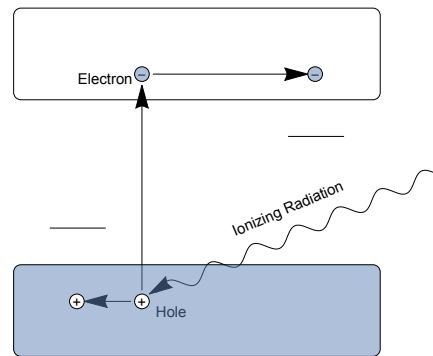
Figure 2.6 shows the four stages of the luminescence process: ground state, ionization, storage, and stimulation. When the crystal is in its ground state, there are no electrons nor holes in the traps within the band gap. When the crystal absorbs radiation, electrons will ionize into the conduction band, leaving behind a hole in the valence band. While in these energy bands, the electrons and holes are allowed to freely travel throughout the crystal. When the charge carriers encounter a trap of the proper type, they will be held in place until they receive enough energy to re-ionize. Electrons that become trapped can be stimulated out of their traps via heat (TL) or photons (OSL), allowing them to recombine with holes. Upon recombination, the electron will relax back to the ground state and release its stored energy in the form of a photon - the luminescence signal. When the majority of electrons have recombined, the crystal returns to its ground state and is considered “reset.” Crystals that have absorbed more dose before resetting will have more electrons in traps and thus will provide more luminescence during stimulation. An example of an OSL measurement is shown in figure 2.7. The luminescence signal decreases exponentially, and the measurement continues until the signal is fully reset, leaving only background counts.

2.2.1 Practical Considerations of Electron Trap Energy Levels

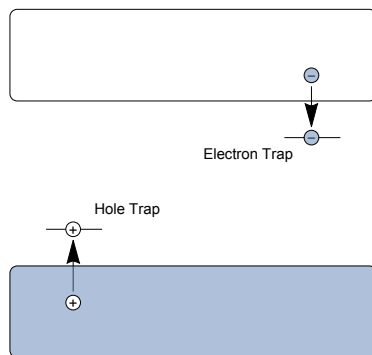
The “trap depth” is the energy level of the electron trap relative to the conduction band, and the energy of the stimulating medium must be greater than or equal to that of the trap



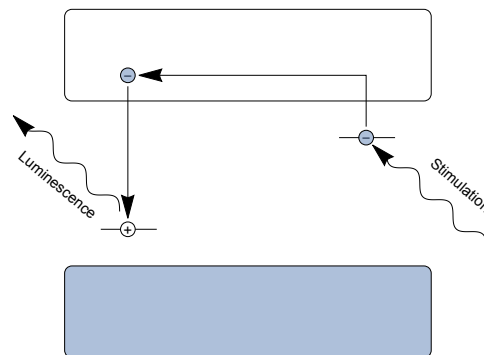
(a) Crystal Ground State



(b) Charge Carrier Ionization



(c) Charge Carrier Storage



(d) Recombination and Stimulation of Luminescence

Figure 2.6: The four stages of the luminescence process. a) the crystal begins in the ground state with electrons completely occupying the valence band. b) When an electron absorbs ionizing radiation, it becomes excited to the conduction band where it is free to travel. c) The electron, and the hole it has left behind, travel within their bands until recombining or becoming trapped. d) Upon thermal or optical stimulation, the electron is ejected from its trap where it will again travel freely through the conduction band. When a traveling electron recombines with a trapped hole, energy is released as a photon and the crystal returns to ground state.

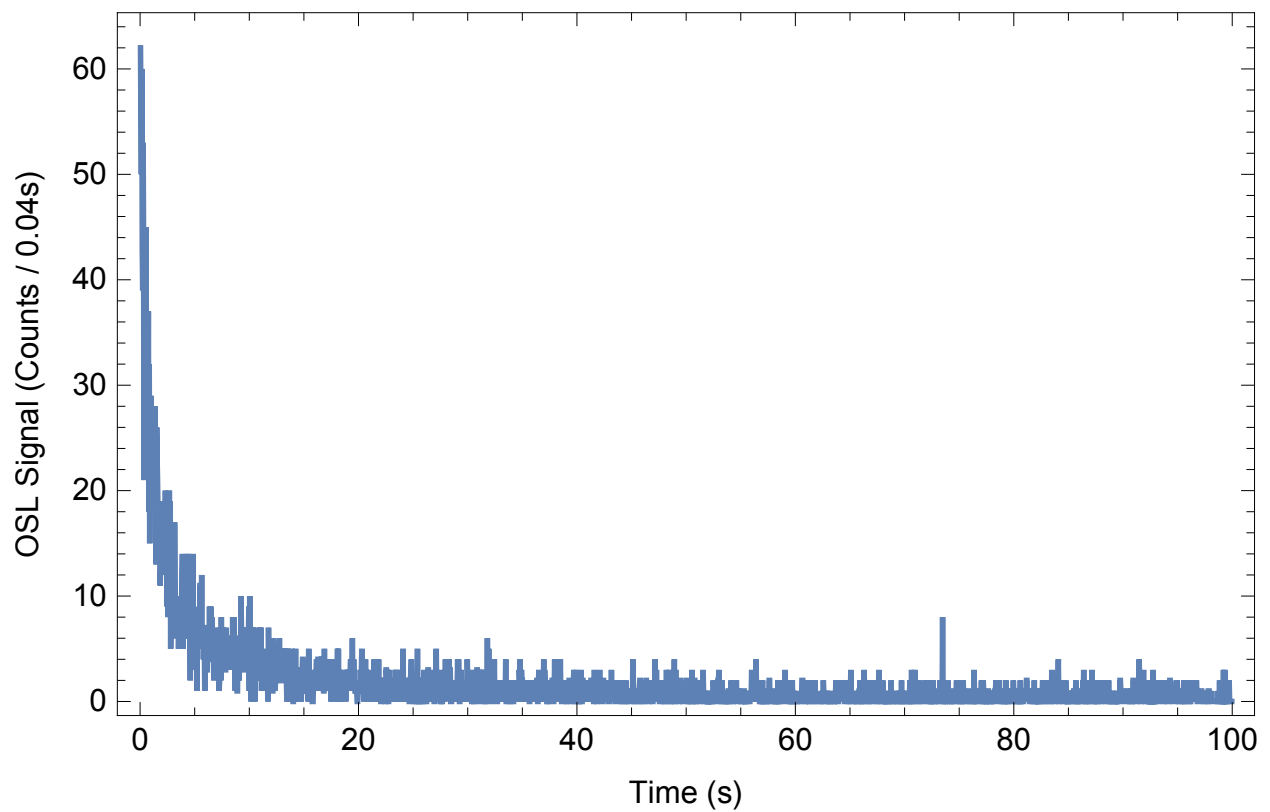


Figure 2.7: Plot of an OSL signal measurement. Only the first few seconds of OSL provide strong signal, but stimulation continues until effective resetting occurs.

depth in order to stimulate an electron from its trap. Different materials produce traps with different depths, driving the need for various optical wavelengths and stimulating temperatures depending on sample and measurement type. Figure 2.8 shows an example thermoluminescence curve measured from three different samples. As the temperature increases, electrons are stimulated from traps at different energy depths.

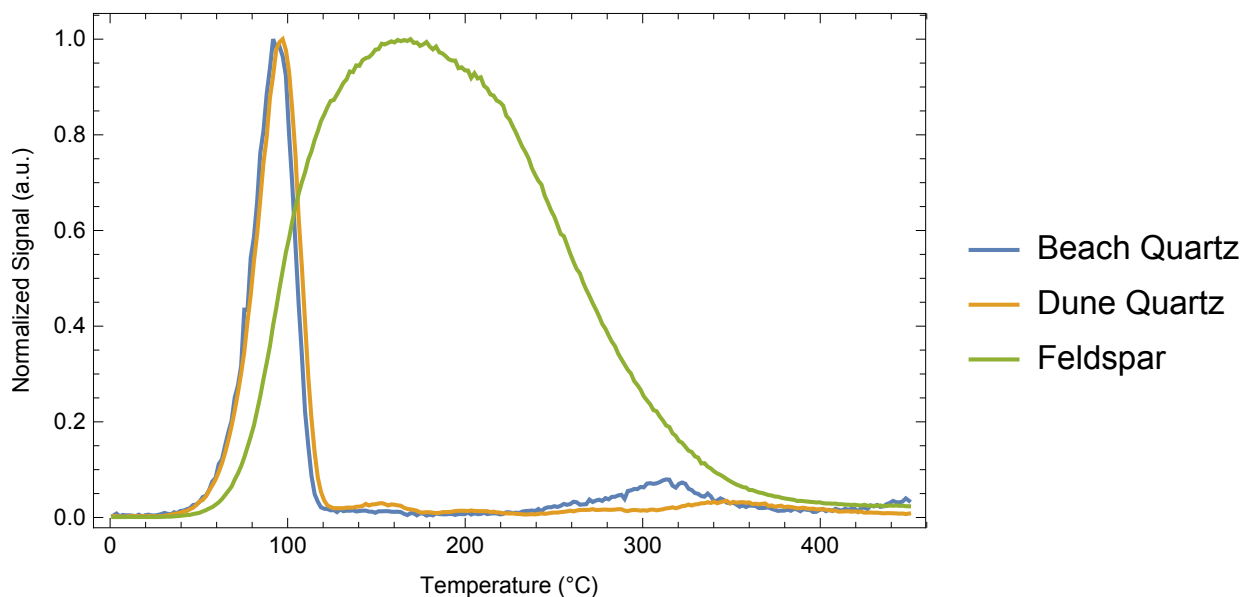


Figure 2.8: Luminescence is only stimulated beyond certain temperatures depending on the energy required by the trap. The quartz data are vastly different from the feldspar, but even the quartz curves show minor changes in luminescence depending on temperature and thus, trap depth. All three samples appear to be fully reset at 400°C.

It can clearly be seen in figure 2.8 that different materials have different trap depths, but even similar materials may have slight variations in their average trap depth. Thus, a more realistic interpretation of the energy band structure of a natural crystal may be found in figure 2.9. Along with the standard OSL electron traps, figure 2.9 adds shallow traps that are easily emptied at room temperature and deep traps that are unable to be stimulated with light.

Energy

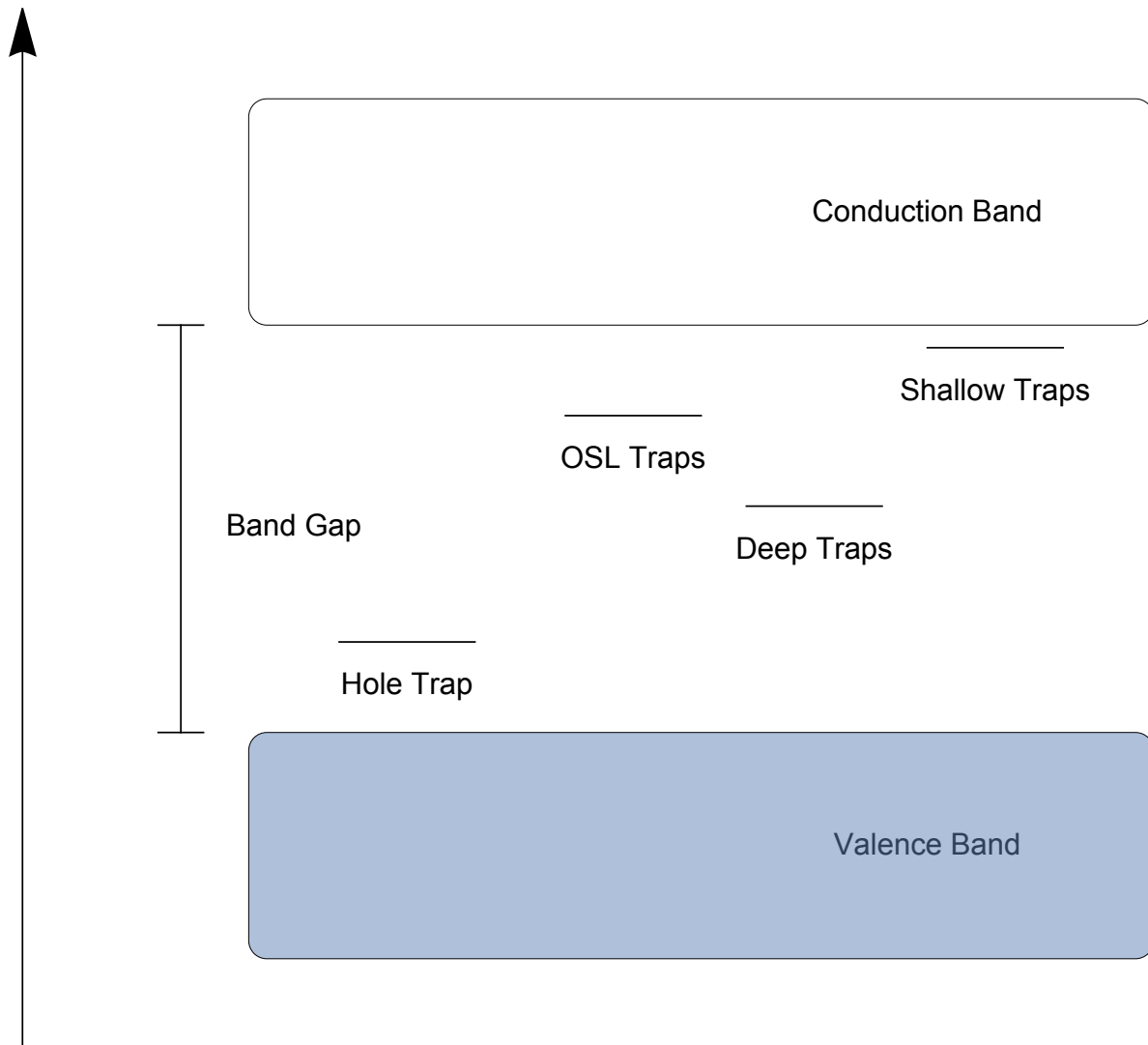


Figure 2.9: In a real crystal, electrons may become trapped in a variety of defects with differing energy levels. Shallow traps are easily emptied at room temperatures while deep traps remain occupied even during optical stimulation. OSL traps are the ones that provide the signal required for luminescence measurements.

2.2.2 Mathematical Description of Luminescence

The simplest model for determining the OSL signal intensity is the one-trap, one-recombination center (OTOR) model (Aitken 1998; Bøtter-Jensen, McKeever, and Wintle 2003; Yukihiro and McKeever 2011). Figure 2.6d is an example of a stimulation event occurring in the OTOR model. The charge neutrality of an OTOR model crystal lattice may be written as a sum of the concentrations of electrons in the conduction band (n_c) and in electron traps (n) being equal to the sum of the holes in the valence band (m_v) and in hole traps (m):

$$n_c + n = m_v + m \quad (2.8)$$

During stimulation of a sample with trapped charge carriers, there is no transition into the valence band, only recombinations. This results in the charge neutrality equation becoming $n_c + n = m$ during stimulation, with a rate of change during stimulation written as:

$$\frac{dn_c}{dt} = -\frac{dn}{dt} + \frac{dm}{dt} \quad (2.9)$$

The rate of change equations dn/dt and dm/dt are written as:

$$\frac{dn}{dt} = n_c A (N - n) - np \quad (2.10)$$

$$\frac{dm}{dt} = n_c B m \quad (2.11)$$

Where $p = \sigma\phi$ is the rate of stimulation of electrons from the trap in terms of the photon flux ϕ and the photo-ionizational cross section σ . A is the trapping probability, B is the recombination probability, N is the total number of electron traps, and $n(m)$ is the number of occupied electron (hole) traps. Next, an assumption of quasi-equilibrium is applied:

prior to stimulation, most, or all, of the electrons in the conduction band will have migrated into traps such that $dn_c/dt \ll dn/dt, dm/dt$ and $n_c \ll n, m$. This reduces equation 2.9 to:

$$\frac{dm}{dt} = \frac{dn}{dt} \quad (2.12)$$

Here we apply our second assumption for simplicity: electrons will not become re-trapped once evacuated, giving $A = 0$ in equation 2.10. Because the luminescence intensity is proportional to the rate of change of the trapped electrons, we may write it as:

$$I_{OSL} \propto \left| \frac{dn}{dt} \right| = np \quad (2.13)$$

Which solves neatly into:

$$I_{OSL} = n_0 p e^{-tp} = I_0 e^{-tp} \quad (2.14)$$

Where n_0 and $I_0 = n_0 p$ are the initial number of trapped electrons and initial luminescence respectively at time $t = 0$ of stimulation. In reality, luminescence systems rarely behave so well; there are multiple traps that each release electrons at different rates as well as the possibility of re-trapping. If stimulated electrons are allowed to become re-trapped, A will no longer be zero in equation 2.10 and a second-order solution is found:

$$I_{OSL} = I_0 \left(1 - \frac{n_0 p B t}{N A} \right)^{-2} \quad (2.15)$$

with $I_0 = n_0^2 p B / N A$. Analysis using equations 2.14 and 2.15 allow a general order equation to be found (Yukihara and McKeever 2011):

$$I_{OSL} = I_0 \left(1 - \frac{n_0 p B t}{N A} \right)^{b/(1-b)} \quad (2.16)$$

where $I_0 = n_0^b p B / N A$, and b is the kinetic parameter - i.e., $b = 1$ for first order kinetics, $b = 2$ for second order kinetics, etc. Finally, if both multiple electron traps and recombination

sites are allowed, rate of change equations are easily generated through superposition:

$$\frac{dn_i}{dt} = n_i p - n_c A_i (N_i - n_i) \quad (2.17)$$

$$\frac{dm_j}{dt} = n_c B m_j - n_v B (M_j - m_j) \quad (2.18)$$

Here, the i and j subscripts denote the number of traps and recombination centers, respectively. The added term in equation 2.18 results from the addition of multiple recombination centers: n_v is the number of holes in the valence band and M_j is the total number of recombination centers. Even these rate equations are not complete, due to the lack of interaction between traps and the effects of recombinations that don't result in OSL signal (non-radiative transitions). Radiative and non-radiative transitions have probabilities of Γ_R and Γ_{NR} , respectively. The ratio of the probabilities of radiative transitions to total transitions is the luminescence efficiency η :

$$\eta = \frac{\Gamma_R}{\Gamma_R + \Gamma_{NR}} \quad (2.19)$$

In 1939, Gurney and Mott discovered that non-radiative transitions were strongly affected by temperature. As the temperature of an electron increases, so does their vibrational energy. There exists an energy W where the energy curves of vibrational ground state and excited state cross. If an excited electron is heated enough to reach this “activation” energy, it will be able to non-radiatively transition into the ground state by emitting a phonon. The non-radiative transition probability may be written as a function of the activation energy and temperature:

$$\Gamma_{NR} = \Gamma_0 e^{-W/kT} \quad (2.20)$$

This non-radiative transition through phonon emission is called thermal quenching and

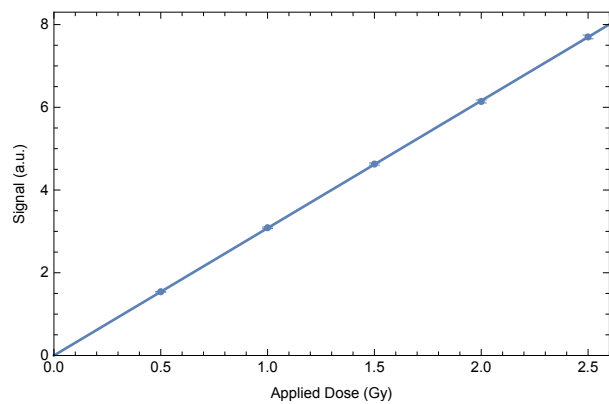
allows the luminescence efficiency to be written as:

$$\eta(T) = \frac{1}{1 + Ke^{-W/kT}} \quad (2.21)$$

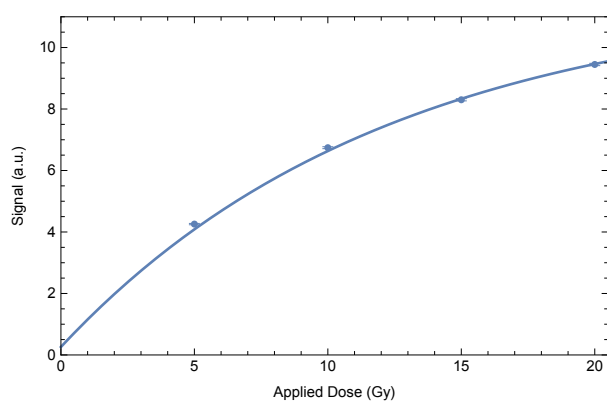
where $K = \Gamma_0/\Gamma_R$. Thermal quenching becomes negligible at sufficiently low temperatures, preventing electrons from performing non-radiative transitions and allowing the luminescence efficiency to go to 1.

2.3 Luminescence as a Dosimeter

Thus far, we have discussed how ionizing radiation causes electrons to become trapped in defects within the crystal lattice, and how optical stimulation causes trapped electrons to emit luminescence. The emitted luminescence is proportional to the number of trapped electrons, which is in turn proportional to the incoming ionizing radiation. Thus, by measuring the OSL signal of a sample, one may determine the amount of energy that a sample has stored from ionizing radiation since its last resetting event. Figures 2.10a and 2.10b show examples of an OSL signal's response to increasing laboratory doses. Figure 2.10a shows linear proportionality of the OSL signal to the applied dose, but in figure 2.10b, it can be seen that samples have a saturation limit to the number of electrons they can store in their traps, resulting in non-linear proportionality at high applied doses. Using these lab-made dose response curves, signal resulting from a natural dose may be measured to determine the sample's equivalent dose – the amount of laboratory-applied radiation required to produce an amount of luminescence that is equivalent to that which was observed during the natural OSL measurement. In the next chapter, we will discuss the conventional instrumentation and methods for performing luminescence dosimetry using OSL.



(a) Linear dose response.



(b) Saturated dose response.

Figure 2.10: The OSL signal's response to the applied dose varies by sample. The response is linear at low dose ranges but becomes exponential as the applied dose reaches saturation.

Chapter 3

OSL for Dosimetry and Dating

In the previous chapter, we discussed how crystals store energy when irradiated, how that energy is emitted as luminescence when stimulated, and how that luminescence may be used to perform OSL dosimetry. Multiple series of commercial instruments are available for luminescence dosimetry, such as the *lexsyg* readers from Freiberg Instruments (Richter, Richter, and Dornich 2013; Richter, Richter, and Dornich 2015), and the more common Risø luminescence readers (Bøtter-Jensen et al. 2000; Bøtter-Jensen et al. 2003). These readers enable the user to perform combinations of luminescence stimulation, luminescence detection, sample irradiation, and sample heating to obtain a reproducible equivalent dose from sediment samples.

The most widely applied procedure for sediment dosimetry is the single aliquot regenerative-dose (SAR) procedure (Murray and Wintle 2000a; Murray and Wintle 2000b). This procedure is regarded as the most reliable measurement procedure for sediment dosimetry, and is also suitable for performing dosimetric dating on rock surfaces. This chapter describes the capabilities of the Risø instrument that was used in this work and serves as an example of a commercial reader for sediment dating. This chapter also outlines the most important steps of the SAR procedure for luminescence dosimetry.

3.1 Risø TL/OSL Reader

The commercially available Risø TL/OSL DA-20 luminescence reader (Fig. 3.1) was used in this dissertation for calibration and characterization purposes (Bøtter-Jensen et al. 2003). Luminescence readers simulate the processes that affect samples in nature, and are thus equipped with a radiation source, facilities for thermal or optical stimulation, a system for signal collection, as well as various software for measurement control and analysis.

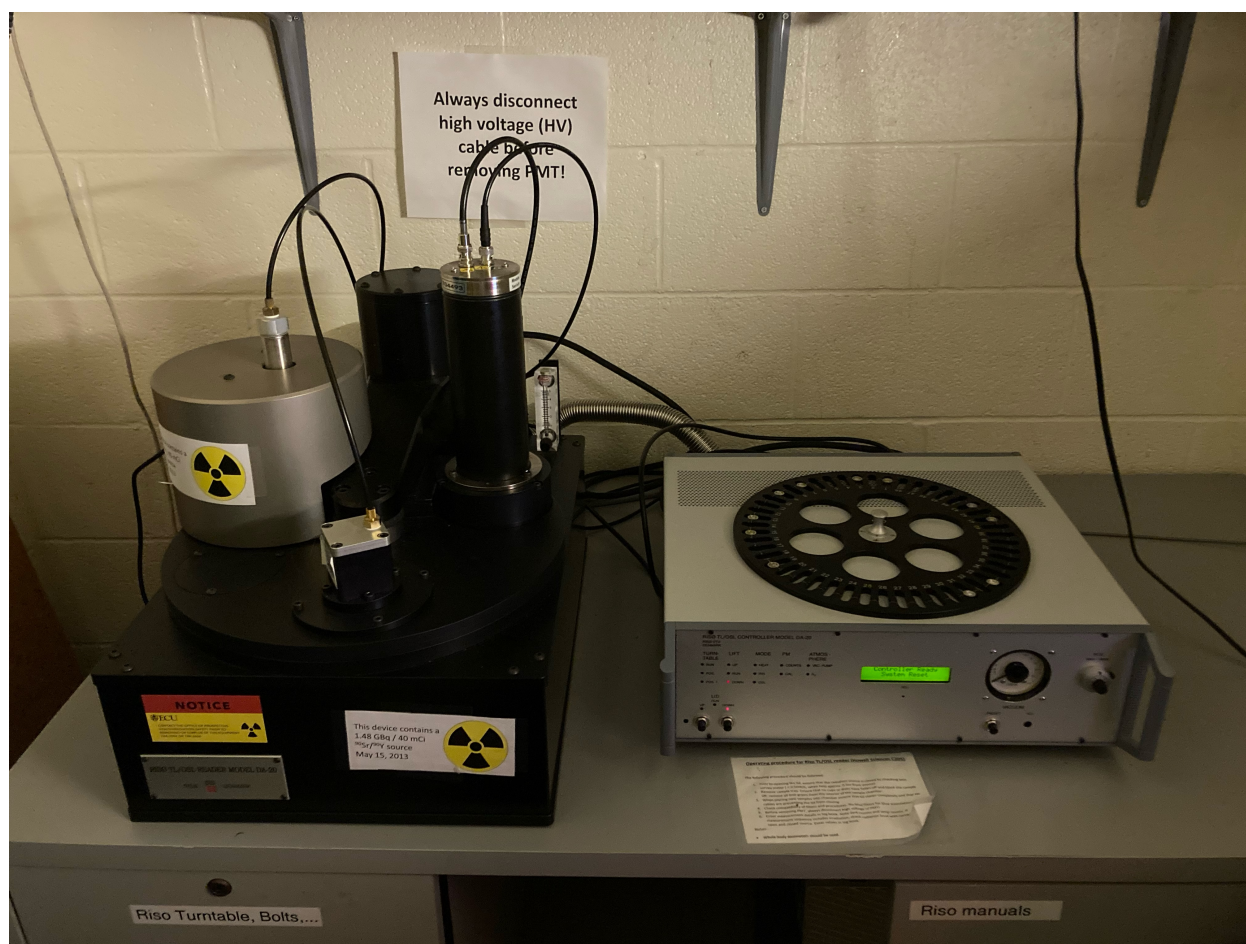


Figure 3.1: Image of the Risø TL/OSL DA-20 luminescence reader (left) and carousel (right). Software controls the measurement process and may perform the full SAR procedure on a number of sample types.

Sediment grains are mounted on the surface of either 9.7 mm diameter stainless-steel disks or 11.65 mm diameter cups (Fig. 3.2) with Rusch® Silkospray™. The disks or

cups are loaded into a 48-slot sample carousel (Fig. 3.1) and placed into the reader. This carousel is also capable of holding small rock slices with a diameter < 12 mm and thicknesses < 1 mm. When the loaded samples are thermally or optically stimulated, their emitted signal is recorded by the luminescence detection system. Using the measurement software, the reader is easily programmable with modular steps to perform any combination of stimulation, heating, and irradiation. This section describes the Risø reader's integrated equipment that is used to perform OSL dosimetry on sediment; the Risø software is described in appendix A.



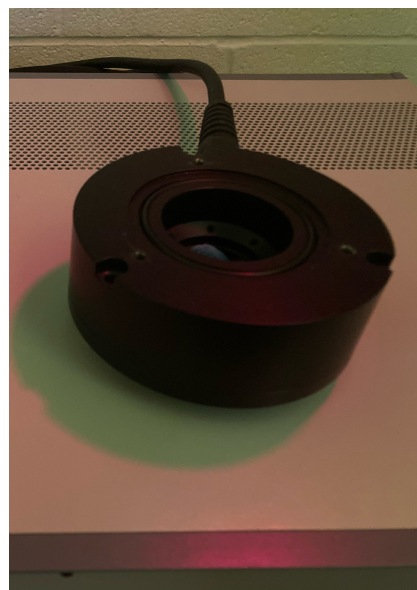
Figure 3.2: Image of the Risø TL/OSL DA-20 luminescence reader cups with eraser for scale. The stainless-steel disks (left) are 9.7 mm in diameter and the stainless-steel cups (right) are 11.65 mm. Sediment is adhered to the disks or cups and placed into the sample carousel for measurement.

3.1.1 Stimulation System

During the measurement sequence, each sample is raised from the carousel to the measurement position by a lift that also acts as a heating element (Fig. 3.3a). The sample is then stimulated by heat and/or a variety of LEDs. It is possible to operate both the heating element and the LEDs at the same time to perform OSL at raised temperatures. The heating element is capable of heating samples up to 700°C at heating rates from 0.1 to 10°C/s. High heating temperatures can cause the element to oxidize, which is prevented by purging the Risø sample compartment with nitrogen gas.



(a) Image of the Risø's heater element. It is able to lift the sample to the measurement area and raise the temperature up to 700°C. The heating element is useful for both low temperature heating during optical stimulation as well as high temperature thermoluminescence stimulation.



(b) Image of one set of modular LEDs for the Risø reader. The LED's have built in emission filters and can be switched depending on which stimulating wavelength is required.

Figure 3.3: Images of the Risø reader's stimulation equipment.

The Risø reader is equipped with modular external LEDs that may be exchanged to measure different sample types (Fig. 3.3b). Available LED arrays include a combined IR (870 ± 40 nm, 145 mW/cm²) and blue (470 ± 20 nm, 80 mW/cm²) array, as well as a green (525 ± 30 nm, 40 mW/cm²) array. Emission filters are placed in front of the LEDs to eliminate short-wavelength emission tails that would interfere with the luminescence

signal: a GG-420 long-pass filter is applied to the blue LEDs, the green LEDs use a GG-475 long-pass filter, and the IR LEDs use an RG-780 long-pass filter. Emission spectra of the LEDs and the transmission spectra of the associated filters are shown in figure 3.4.

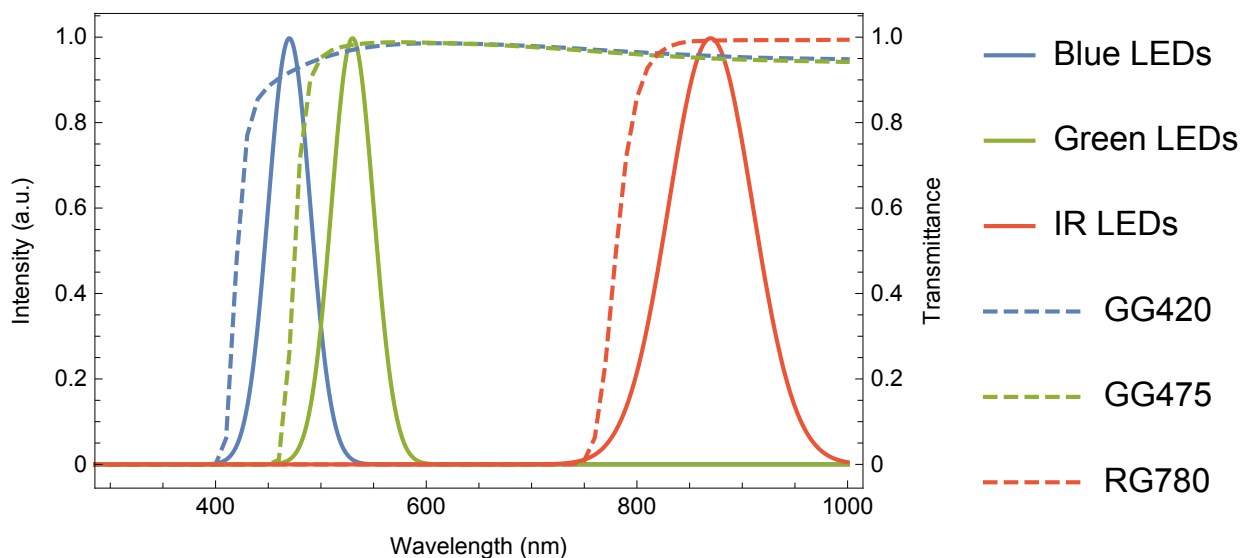


Figure 3.4: Wavelengths for the blue, green, and IR LEDs and transmittances for their respective filters.

3.1.2 Luminescence Detection System

To detect the luminescence signal, the Risø reader contains a photomultiplier tube (PMT) with manually changeable detection filters. The filters are used to prevent the longer-wavelength stimulation light from reaching the PMT and pass only the shorter-wavelength luminescence. The emission spectra of the LEDs and the transmission spectra of the luminescence-passing detection filters used in this dissertation are described in appendix E.3.2. The detection filters are selected depending on sample composition and stimulation wavelength. For example, quartz must be stimulated with blue or green light to obtain a usable signal, which is emitted in the UV range. When stimulating quartz with the blue LEDs, the detection filter must block the blue range and pass the UV range. When necessary, apertures of different sizes are added to prevent exceptionally bright samples

from saturating the PMT.

3.1.3 Radiation Source

To calibrate the sample's dose response, the Risø reader is equipped with a shielded $^{90}\text{Sr}/^{90}\text{Y}$ beta radiation source. The dose rate of the beta source is proportional to $1/r^2$ and decreases drastically with distance. However, at closer distances, spatial variations in the dose rate become more pronounced leading to decreased dose rate uniformity. The source-to-sample distance in the reader is 7 mm, balancing spatial uniformity with dose rate. When activated, the source window takes 0.11 s to open, and this shutter time is subtracted from the programmed irradiation time to correct for the delay. Due to the proximity of each carousel slot with its neighbor, there is an irradiative “cross-talk” of 0.250 ± 0.003 % of the given dose to adjacent slots and only 0.014 ± 0.002 % to the second nearest slot (Thomsen et al. 2006). Because of this, every other carousel slot is left empty to minimize the effects of cross-talk, allowing up to 24 samples to be measured at a time.

On December 17, 2013, the $^{90}\text{Sr}/^{90}\text{Y}$ beta source was calibrated by Dr. Regina DeWitt for quartz sediment using a ^{60}Co source, resulting in a dose rate of 126.2 ± 3.9 mGy/s. This dose rate has been calculated to have fallen to 103.0 ± 3.2 mGy/s as of June 1, 2023, due to the half-life of the source. The beta source was also previously calibrated by Emmanuel Osunkwor for Luxel[®]+ dosimeter sheets on April 23, 2021, with a dose rate of 72.7 ± 1.5 mGy/s by using a ^{137}Cs source (Osunkwor 2022). This research calibrated the Risø's beta source against a ^{60}Co source in March 2023 for quartz sediment as well as quartzite and sandstone rock slices. These recent calibration measurements and their results are discussed in chapter 7.

3.2 Luminescence Dosimetry Measurements

The luminescence emitted by a sample is directly correlated with the sample's absorbed dose; however, the mathematical relationship between signal and dose varies by sample and must be calibrated. To this end, a dose response curve is built (Fig. 3.5). First, the signal resulting from the unknown natural dose is measured. Then, the sample is irradiated in the laboratory with increasing doses and a curve is fit to the resulting measured signals. Once this curve has been generated, the “natural” signal is superimposed onto the fit and the natural equivalent dose D_e is found. While the basis of the dose response is straight-forward, the practical measurement process is more complicated. This section outlines the conventional SAR measurement procedure used for sediment dating which will be adapted for the dating of rock surfaces.

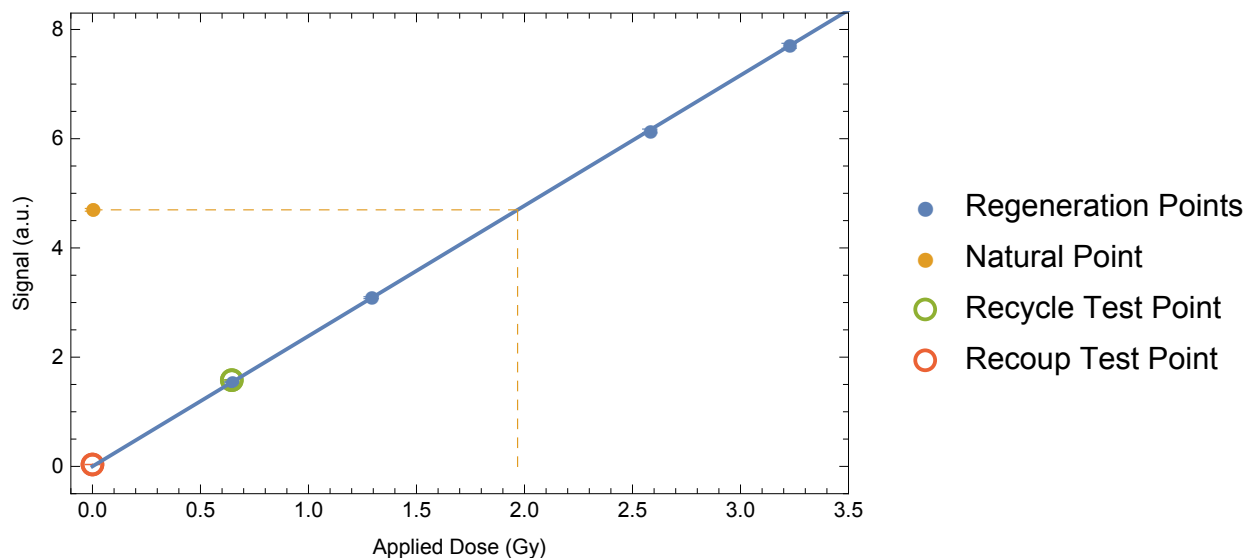


Figure 3.5: A dose response curve featuring four laboratory dose points, two test points and the natural point. The two test points are overlapped by the first two laboratory dose points, which indicates that the sample passes all tests. The natural point is in orange, plotted on the y-axis, and interpolated onto the fit curve. The luminescence for each point is measured as a ratio of the OSL signal divided by a test signal, provided by a test dose. The measurement methods are described in detail throughout this section.

3.2.1 Effects of Shallow Traps and Repeated Irradiations on the Luminescence Signal

As shown in figure 2.9, natural materials have a variety of different traps with varying luminescence properties. Some of these traps have properties that have the potential to negatively affect the dose measurement. Shallow traps are electron traps that are close enough to the conduction band to be easily evacuated at room temperature. These traps will empty over larger timescales (hours to days), but fill up during laboratory irradiations, leading to inflated laboratory signals relative to the natural signal. As a result, a lower natural dose is measured unless these traps are accounted for. To correct for their effect, the SAR procedure adds a low temperature (160-300°C) “preheat” step after irradiation to empty the shallow traps without stimulating the primary luminescence traps. This preheat temperature must be determined on a sample-by-sample basis, as each sample will have varying defects and trap depths.

Thermal quenching (see section 2.2.2) leads to a decrease in luminescence efficiency with increased temperature; however, electrons that are detrapped during stimulation can become retrapped in shallow traps at room temperature, also decreasing luminescence efficiency. OSL stimulation is therefore carried out at a temperature that is higher than that of the thermal stability of the shallow traps, reducing the shallow traps’ ability to retain energized electrons and increasing the overall emitted luminescence.

Deep traps are those that are not stimulated upon light exposure and will fill up over repeated cycles of irradiation and measurement. As fewer deep traps are available, more electrons will become trapped in radiative traps, even if the same dose is given. Repeated irradiations can therefore cause an increase in luminescence sensitivity in samples (Fig. 3.6a). To monitor this sensitivity change, a small test dose is applied after every measurement and the test signal is read after a modified preheat step called the “cutheat.” Each measurement signal is divided by its test signal to generate a point on the dose response

curve (Fig. 3.6b).

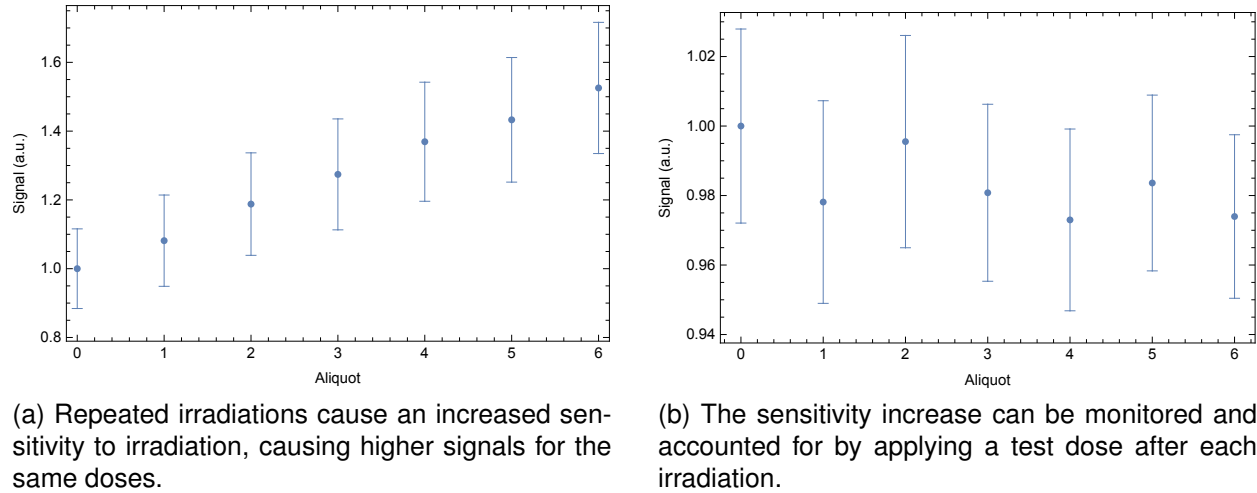


Figure 3.6: A constant dose of 450 seconds of beta irradiation was applied. Figure (a) had no test dose correction, while figure (b) had a test dose of 50 seconds after each main irradiation.

3.2.2 Single Aliquot Regenerative-Dose Procedure

The single aliquot regenerative-dose (SAR) procedure outlined in table 3.1 measures a sample's dose by combining repetitions of irradiations and measurements with built in tests for preheat temperature and luminescence sensitivity changes (Murray and Roberts 1998; Murray and Wintle 2000a; Murray and Wintle 2000b; Murray and Wintle 2003). A sample is split into multiple subsamples called “aliquots,” which are passed through multiple measurement cycles indexed by i . Before the first cycle is run, the expected natural dose of the sample is crudely estimated by measuring a single aliquot's natural signal and comparing it to a laboratory generated signal. This allows us to choose laboratory doses that encompass the expected natural dose. The first cycle, in which the natural signal is measured, has the index $i = 0$, and no laboratory dose is administered ($D_0 = 0$ Gy). Each following cycle starts with an applied dose of D_i and is followed by a preheat step that is held for 10 seconds to empty shallow traps. Next, the OSL signal (L_i) is measured for 100 seconds at 125°C – long enough to thoroughly reset the sample (Fig. 2.7). A constant

test-dose (D_t) that is set to be $\lesssim 20\%$ of the expected natural dose is administered before a cutheat at 160°C is applied. Finally, the test OSL signal (T_i) is measured and a “corrected” signal is found by dividing the initial OSL signal L_i by the test signal T_i . Plotting the relative signal L_i/T_i against the laboratory dose produces a “regeneration point” for the dose response curve. One natural point and four to five regeneration points are sufficient to recover an accurate natural absorbed dose. The SAR procedure includes additional cycles that test the reliability of the procedure. These additional tests are described in detail in appendix B.

Table 3.1: Steps of an SAR cycle. Cycles are indexed by i , where $i = 0$ is the initial natural cycle and $D_0 = 0$ Gy. For each regeneration and test point, $i = 1, 2, \dots, n$ and the constant test dose (D_t) will give a test signal for all i .

Step	Action	Associated Value
1	Irradiate with a dose	D_i
2	Preheat for 10s	-
3	Measure OSL for 100s	L_i
4	Irradiate with test dose	D_t
5	Cutheat to 160°C	-
6	Measure OSL for 100s	T_i
7	Repeat steps 1-6 for each regeneration and test point	-

3.3 Ideal Samples

An ideal sample for OSL dating is bright, thermally stable over geological timescales, and has been completely reset by the event to be dated. Sediments made up of quartz (SiO_2) and potassium-rich feldspar minerals form the bulk of conventional samples used for luminescence dating. Sediment grains ($<250\ \mu\text{m}$) are easily transportable, providing opportunities to date a variety of transportation events (aeolian, fluvial, glacial, etc.), and they are easily resettable owing to the fact that sunlight stimulates luminescence through-

out the entirety of the grain. Excepting alpha radiation, sediments of this size experience a uniform, or well-characterized, dose flux, and the alpha-affected portion is easily removed during sample preparation using etching with hydrofluoric acid. During measurement, sediment grains tend to be bright, even at low doses, and have a sensitive response to increasing dose. Usable sediment grains are omnipresent across the planet, save for a few locations, allowing one to find *something* to measure in most desirable sites. However, when in sediment-barren locations like the Antarctic Peninsula, or when dating structures built with stones, one must look to the less conventional cobble surface for dating. The next chapter will discuss the limitations of sediment methodologies when applied to rock surfaces.

Chapter 4

Considerations and Requirements for Spatially Resolved Measurements

In the previous chapter, we discussed the instrumentation and methods for conventional dosimetry and dating. It was explained that sediment samples are generally preferable to cobbles for optically stimulated luminescence (OSL) dating, but suitable sediments are not always available on-site. For example, the few sediments in the windy environment of the Antarctic Peninsula are continually transported and reset while cobbles tend to stay in place and retain OSL-relevant age information (Simms et al. 2011; Simkins, Simms, and DeWitt 2013). Rock surfaces have also been used to date the construction or destruction of buildings (Liritzis 1994; Greilich, Glasmacher, and Wagner 2005; Vafiadou, Murray, and Liritzis 2007). This makes cobbles the more robust choice for OSL dating in these sites but, as we will see, dating cobbles with conventional instruments is fraught with challenges. In this chapter I will discuss the requirements for applying the single aliquot regenerative-dose (SAR) procedure to cobble surfaces. I will show that the commercially available luminescence readers are not well suited to this task and that the dose must be measured with spatial resolution. Lastly, I will discuss the basic components and general setup of an instrument that enables the measurement of a spatially resolved

“dose map.”

4.1 Challenges of Cobble Surface Dating

Unlike cobbles, sunlight can penetrate through an entire sediment grain and the trapped charges responsible for OSL are removed (i.e. “reset”) by sunlight. For cobbles, there is a maximum depth that sunlight can penetrate to bleach the cobble’s signal. In order to obtain the portion of a rock that has been fully reset, the outer 1 mm of the cobble must be isolated using rock cutting and slicing techniques (Simms et al. 2011; Simkins, Simms, and DeWitt 2013).

Technically, it is possible to place intact, small, and thin rock slices into the Risø reader (diameter <12 mm and thickness 1 mm). However, due to the nature of the Risø reader, the entire slice is stimulated, and the integrated signal is detected. Rocks are inherently non-uniform, consisting of a multitude of minerals and grain sizes, which leads to large uncertainties when a whole slice is measured. These non-uniformities mainly develop in four ways: 1) Different minerals, such as quartz and feldspar, have different luminescence properties so they must be stimulated at different wavelengths while they emit luminescence in different wavelength bands. 2) K-feldspars have internal radioactive ^{40}K , while quartz is considered inert. Because of feldspar’s added internal component, quartz and feldspar grains within a rock are exposed to different dose rates. Feldspar’s internal radioactivity is also grain size dependent. As a result of these facts, these two types of minerals, as well as minerals of different grain sizes, will have different doses, even if they were reset by the same event. 3) Rocks contain a larger fraction of uranium- and thorium-bearing heavy minerals that lead to large dose-rate non-uniformities. Therefore, the dose absorbed by a specific grain strongly depends on its exact position within the rock and its nearest mineral neighbors. 4) Lastly, the heavy minerals are often opaque and are not easily reset by sunlight but emit a bright luminescence signal. Due to these

four factors, the dose measured from a whole rock slice using a Risø reader would be an average value of the dose values resulting from these non-uniformities.

Sediment samples also experience these non-uniformities. However, they can be subjected to elaborate sample preparation treatments to obtain pure mineral extracts (quartz or feldspar) of a defined grain size range (Rhodes 2012). To be measured with the same Risø instrument and the same methods as sediments, rock slices must be ground into sediment-sized grains and prepared in the same way. The process of cutting, slicing, and grinding rock samples complicates the dating process fourfold: 1) With the current technology and technique of rock cutting, slicing only the outer 1 mm of a rough cobble sample is difficult, time consuming, and prone to error, 2) The grains of the rock begin to break during grinding, and there is no way to know the true size of the grains – information which is required to calculate the dose rate for dating, 3) There is a risk that the slicing and grinding process imparts enough energy in the form of heat and pressure to stimulate luminescence and affect the recovered equivalent dose of the sample (Bateman et al. 2012), and 4) While grinding the grains into sediment necessarily dislodges them from one another, it also destroys the spatial information inherent in the cobble as well as any information about the radiation environment experienced by the individual grains.

It is the desire to record the spatial information associated with the stored dose that spurred the subject of this research: the creation of a novel Luminescence instrument with Confocal and Imaging unit for Dating and Dosimetry (LuCIDD). Measuring whole cobbles not only maintains the grain size and spatial information, but also improves the accuracy of the results while simultaneously reducing the time and work required to prepare samples. To overcome the challenges mentioned above, LuCIDD has been built, characterized, and tested to create dose maps of cobble surfaces by stimulating and recording luminescence from small points (pixels) on the surfaces and calibrating those signals using known laboratory irradiations. Beyond the application to whole rock surfaces, LuCIDD's dose maps will also allow characterization of non-uniform radiation fields, specifically in the realm of

medical dosimetry.

4.2 Considerations of Building a Dose Map

As explained in Chapter 2, all luminescence instruments must have the basic components for stimulation and detection of luminescence: a light source, a heat source, and a luminescence detector. The biggest difference between a conventional instrument and an instrument for dose mapping is the need to measure signal data from spatially-resolved pixels on the surface of the cobble. Previous research has accomplished spatially-resolved measurements in two ways: 1) a CCD camera is used to measure the signal from an entire surface during a single stimulation (Duller, Bøtter-Jensen, and Markey 1997; Greilich, Glasmacher, and Wagner 2002; Thomsen et al. 2018), or 2) a laser is used to stimulate points on the surface, and the resulting signal is captured one pixel at a time (Duller et al. 1999; Jacobs et al. 2013).

Both of these measurement methods have merit. The CCD method allows measurement of the entire sample with only a single stimulation measurement. This provides for faster measurements but only allows stimulation and detection of a single wavelength. Using lasers would slow down measurements but would theoretically allow the use of different stimulation and detection wavelengths for measuring different individual mineral grains. Lasers are also much higher in intensity than standard LEDs and will allow for much faster individual measurements even if surface measurements take longer as a whole. The latter approach was chosen for LuCIDD.

Luminescence instruments require stimulation and detection facilities but must also be equipped with a known laboratory radiation source to calibrate the signal and provide equivalent dose information. While the Risø reader used in this research is equipped with a radioactive beta source, X-ray sources are also a valid option for irradiation (Andersen, Bøtter-Jensen, and Murray 2003; Thomsen et al. 2006). Sections 4.3, 4.4, and 4.5

discuss considerations associated with the stimulation, detection, and irradiation facilities capable of developing dose maps of sample surfaces.

4.3 Stimulating Luminescence from Pixels

It is possible to purchase an attachment for the Risø reader that allows measurements of single grains using laser stimulation. This attachment uses a 10 mW green (532 nm) laser to stimulate grains placed on a 10 by 10 grid. The grains are placed into 300 μm deep sample holes which are separated by 600 μm , minimizing the cross-talk of the stimulating laser. Previous research has characterized the Risø single grain laser stimulation size and power to perform SAR procedures on single grains (Duller et al. 1999). It was found that a focal spot of 120 μm was attainable and produced a high power density of $>50 \text{ W/cm}^2$ at each grain, requiring a stimulation time of only 2.2 seconds.

In the single-grain readers, individual grains are placed in separate sample holes. Rock surfaces are continuous, and the grains directly touch their neighbors. The determining factors of pixel size, and thus dose map resolution, is therefore the focal spot of the stimulating laser. Laser power determines the duration of the measurement. Lasers with lower power must stimulate the sample for a longer time to properly bleach the pixel and create reproducible results. Likewise, increasing the stimulation power enables shorter stimulation times; this is especially useful for reducing measurement times when taking multiple datapoints over the surface of a sample.

4.3.1 Beam Size

The power density of a laser is a simple division of the total applied laser power by the area of the focal spot of the laser. This value increases inversely with the square of the focal spot size. Determining and minimizing the focal spot of the laser requires an understanding of gaussian beams and beam expansion which is summarized here and

can be found in more detail in Hecht (2017).

Gaussian Beams

All four of LuCIDD's lasers have gaussian profiles (Fig. 4.1a): their intensities fall off along a distance r radial from the central axis of their respective beams (Fig. 4.1b). It is standard to assign an arbitrary boundary to define the half-width of the beam ($r = w$) where the electric field drops from E_0 to E_0/e . This allows us to write the intensity of each beam:

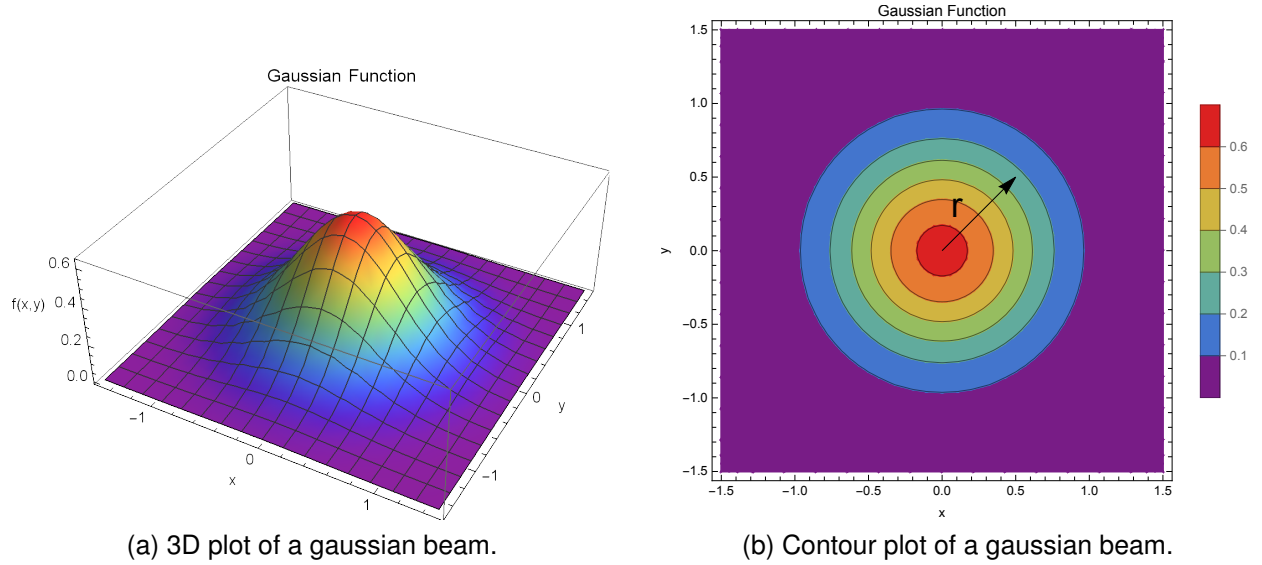


Figure 4.1: Example of a gaussian profile. In (b), r is the distance measured away from the central axis in the transverse plane.

$$I = I_0 e^{-2r^2/w^2} \quad (4.1)$$

where our arbitrary boundary of $r = w$ gives $I = I_0/e^2 \sim 13.5\%I_0$. Due to the geometry of the resonant cavity which produces the laser beam, the beam may become slightly “focused” in the center which produces a minimum beam radius, or beam waist w_0 with divergence Θ (Fig. 4.2):

$$\begin{aligned} w_0 &= \sqrt{\frac{\lambda L}{2\pi}} \\ \Theta &= \frac{2\lambda}{\pi w_0} \end{aligned} \quad (4.2)$$

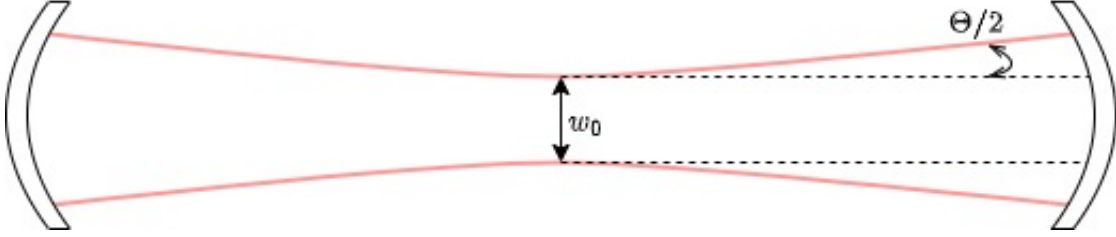


Figure 4.2: Example of a diverging beam inside of a reflective cavity with beam waist w_0 and divergence Θ .

where L is the length of the resonant cavity and λ is the wavelength produced by stimulated emission inside the cavity. Thus, a laser with a smaller beam waist will have a greater divergence. Because of this, it is useful to expand the beam to reduce divergence over long laser paths.

Beam Expansion

A beam expander is a combination of optical components used to increase the diameter of a collimated beam. Figure 4.3 shows a schematic of a Galilean beam expander which was used in this work. Increasing the diameter of a laser will reduce its power density as well as its divergence, leading to smaller beam sizes over longer beam paths. As the beam is expanded, its area is increased quadratically relative to the magnification of the beam expander, reducing the power by a factor of $1/r^2$.

For both telescopes and beam expanders, their magnification (M) may be written as the ratio of their component lens focal lengths where f_o is the objective lens and f_i is the image lens:

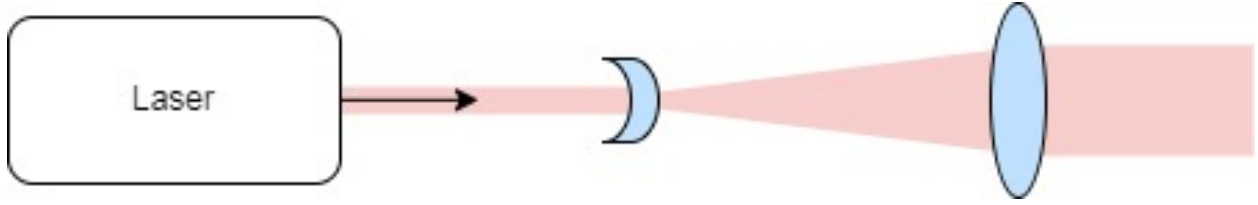


Figure 4.3: Schematic of a Galilean beam expander.

$$M = -f_i/f_o \quad (4.3)$$

Magnification may also be expressed in terms of beam divergence and beam diameter:

$$M = \Theta_o/\Theta_i = D_i/D_o \quad (4.4)$$

Where Θ_o is the divergence of the output beam, Θ_i is the divergence of the input beam, D_i is the diameter of the input beam, and D_o is the diameter of the output beam. For any beam, the diameter of the beam D_L at a path length L is easily determined:

$$D_L = D + L \tan(\Theta) \quad (4.5)$$

Here, the $L \tan(\Theta)$ term dominates the equation at large L so any decrease in Θ greatly affects the beam diameter at long path distances (Fig. 4.4).

4.4 Luminescence Detection

Using highly focused lasers for stimulation poses a challenge; they are more powerful than natural sunlight and can stimulate luminescence below the natural bleaching depth, leading to “pollution” of the natural signal. Research performed by Simms et. al. (2011) explored the effects of sunlight exposure on the surface of cobbles. A granite cobble was cut into 1 mm slices and irradiated with 70 Gy. The stack of slices was bleached from the top with sunlight for an hour. The slices were then prepared similar to sediments. These

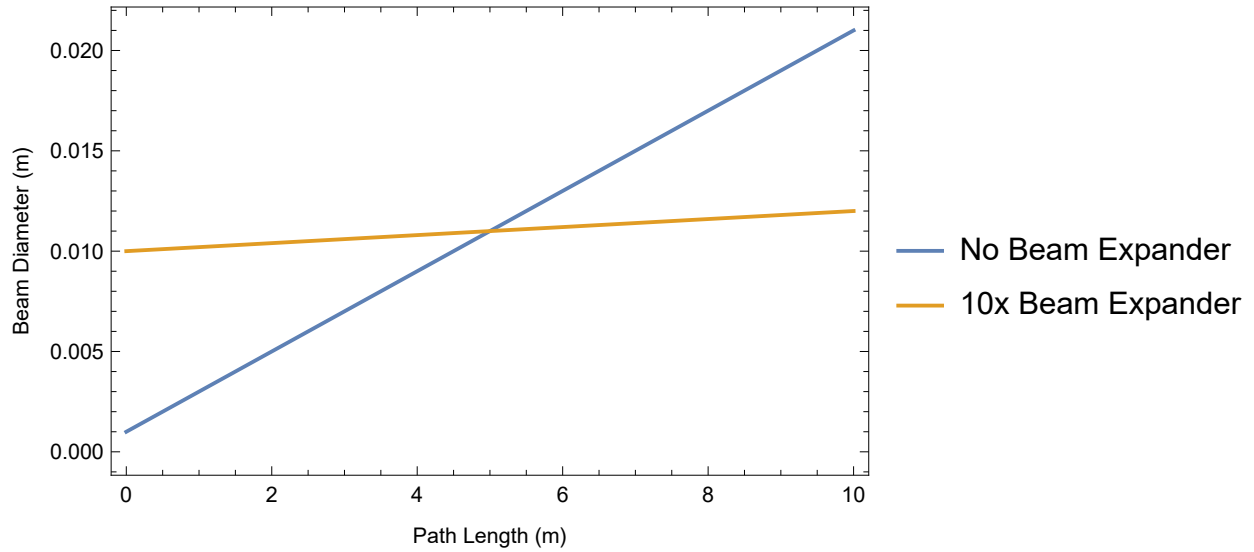


Figure 4.4: Beam diameter versus path length for beams with and without a 10x beam expander with $D_i = 1$ mm, $\theta_i = 1$ mrad, and $M = 10x$. Magnification of a beam causes its divergence to decrease such that an expanded beam will be smaller over long distances than one without expansion.

preparation methods are detailed in section 4.1 to obtain quartz separates. The signal was measured after bleaching and again after a 10 Gy test dose. The bleached signal was normalized to the test dose signal, and it was found that the OSL signal in the upper 1 mm of a cobble's surface had been depleted to 13%.

In 2022, Fuhrmann et. al. expanded on this research with a longer bleaching time and exploration of bleaching of IRSL traps as well as OSL traps. Prior to bleaching, the outer 5 cm was removed from multiple rock samples to ensure that the areas being investigated had never been bleached before. The whole cobbles were placed onto the roof in direct sunlight to be bleached for 108 days. After bleaching, the samples were cored and sliced at 0.85 mm increments and measured as whole slices using a luminescence reader. It was found that the OSL traps of the samples had been bleached to depths of around 4 mm, but the IRSL traps had only been bleached to 2.5 mm. Polluted signals arise when samples experience laser stimulation beyond the depth of their bleached traps, generating large errors in the equivalent dose measurement. Thus, it is desirable to filter out any luminescence that originates below the surface of the sample. Confocal microscopes

have been used for decades to reject signal outside of the focal plane of the objective, making these microscopes a possible solution to the problem of signal pollution.

4.4.1 Confocal Microscopy

In standard microscopy, light is passed through a thin sample located at the focal plane and collected by an objective lens. For thick samples, light originating from planes above and below the focal plane are also collected, resulting in a blurry, low-resolution image. The main purpose of confocal microscopy is to use a diffraction-limited laser spot to illuminate the sample while rejecting out-of-focus light using a pinhole aperture, greatly enhancing the resolution of images obtained from thick samples (Pawley 2006). Figure 4.7 shows a schematic of a confocal microscope that can be used to discriminate collected signal based on the z-axis position of the origin of the signal. The laser is collimated and reflected through an objective by a dichroic mirror that reflects the laser light and allows longer wavelengths to pass. Although the stimulation light is predominantly focused in the focal point of the objective, light scattering leads to a larger stimulated volume. The entire stimulated volume, including out-of-focus areas, will emit luminescence which will travel back through the objective and pass through the dichroic mirror. Beyond the dichroic mirror is a pinhole, located in the focal point of the objective. Since both the pinhole and the stimulated point on the sample are in the focal points of the objective – hence the name “confocal” – the pinhole blocks light from the out-of-focus planes. The transmitted light will strike the detector and become the collected signal.

A confocal microscope may only image a single point at a time, requiring numerous measurements to create a full image. An image is comprised of a grid of pixels; the scanning technique implemented for confocal microscopes scans points, or pixels, along one row. The process then performs a carriage return, like a typewriter, and repeats the scan until all rows of pixels in the image have been recorded. The pixels are compiled together in post-processing to create a full image where the size of the pixels depends

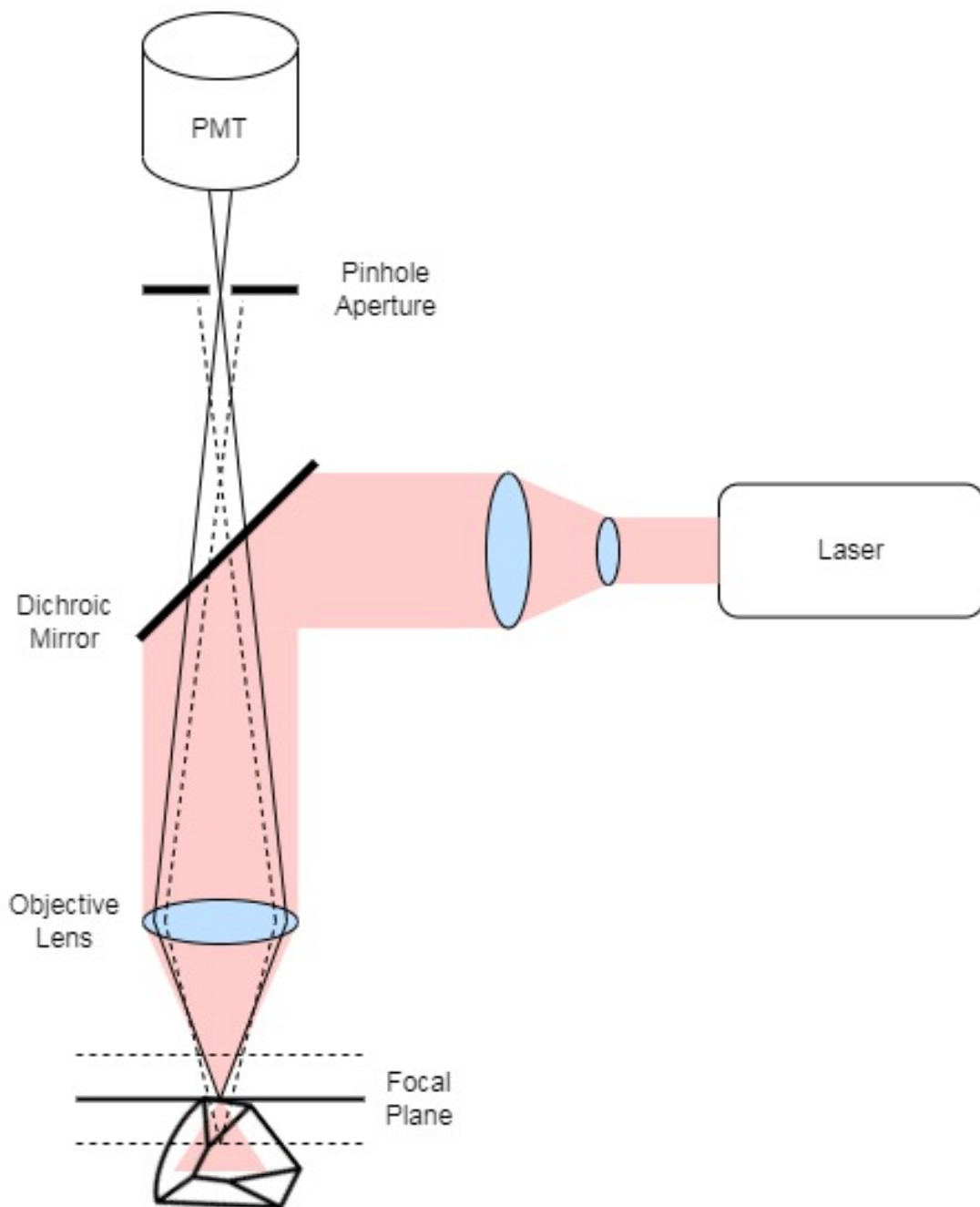


Figure 4.5: Front-view schematic of a confocal microscope. The light from a laser is reflected by a dichroic mirror and passes through the objective as a collimated beam to the sample. The laser stimulates luminescence from the sample and the emitted luminescence travels back through both the objective and the dichroic mirror. When the luminescence reaches the pinhole aperture, the aperture rejects all light that did not originate from the focal plane of the sample. Only luminescence that is in the focal plane of the sample will be measured by the detector. The solid lines represent the path of the in-focus luminescence, while the dotted line represents the path of the out-of-focus luminescence.

on the resolution of the microscope. The resolution in the lateral x-y plane (R) of any microscope is limited by diffraction and is approximated by using the Rayleigh criterion. The Rayleigh criterion states that two points are laterally resolvable if the center of the first point's Airy disk lies at or beyond the first minimum of the second image's Airy disk:

$$R = \frac{0.61\lambda}{NA} \quad (4.6)$$

Where λ is the wavelength of the laser and NA is the numerical aperture of the objective. For standard microscopes to reach this limit, they must be using a diffraction-limited light source and are still not able to resolve image data in the z-axis. For confocal microscopes, R is further refined by applying the detection pinhole. This pinhole reduces the point spread function of the microscope by around 30%, providing an approximate lateral resolution of:

$$R = \frac{0.4\lambda}{NA} \quad (4.7)$$

Note that in order to reach this resolution limit, the pinhole diameter d must be equal to one airy unit which depends on the total magnification of the microscope M :

$$d = \frac{0.61 * M * \lambda}{NA} \quad (4.8)$$

For confocal microscopes that employ detection pinholes equal to one airy unit, the z axis resolution (R_z) is:

$$R_z = \frac{1.4\lambda\eta}{NA^2} \quad (4.9)$$

where η is the index of refraction of the medium between the objective and the sample.

4.4.2 Reflective Objectives

A microscope objective is required for focusing stimulating light when applying confocal microscopy. The selection of objective is an important consideration for measuring luminescence because of the various wavelengths the objective will have to pass. For example, a refractive microscope objective is made of glass and will not pass UV light - the main wavelength of quartz luminescence. Thus, a reflective objective is preferred for its ability to pass all stimulation and luminescence wavelengths unperturbed.

Reflective microscope objectives utilize mirrors instead of lenses to focus light without chromatic aberration and with negligible aberrations of other types. Reflective objectives are built following the Schwarzschild design, consisting of a small convex “secondary” mirror, held in place by a set of spider mount legs, and a larger concave “primary” mirror with an aperture hole in its center (Fig. 4.6). As collimated laser light travels through the aperture hole, it strikes the secondary mirror, reflecting the light to fill the primary mirror. The primary mirror then focuses the beam to a small spot at the focal length of the objective.

Light traveling through reflective microscope objectives experiences obscuration at two points. The first is at the center of the secondary mirror, where the light is reflected directly back through the central aperture of the primary mirror. The second is experienced as the light is focused past the secondary mirror mount and is partially blocked by the spider mount legs. To prevent the light from reflecting back through the aperture, an absorptive coating is applied to the center of the secondary mirror. Due to the effects from both the spider legs and the obscuration spot, total transmission is reduced by 27%.

4.5 Laboratory Irradiation with X-Rays

In-lab irradiations are an important part of the OSL process as they are used to calibrate the dose-to-signal response of a sample. As explained in chapter 3, the SAR dat-

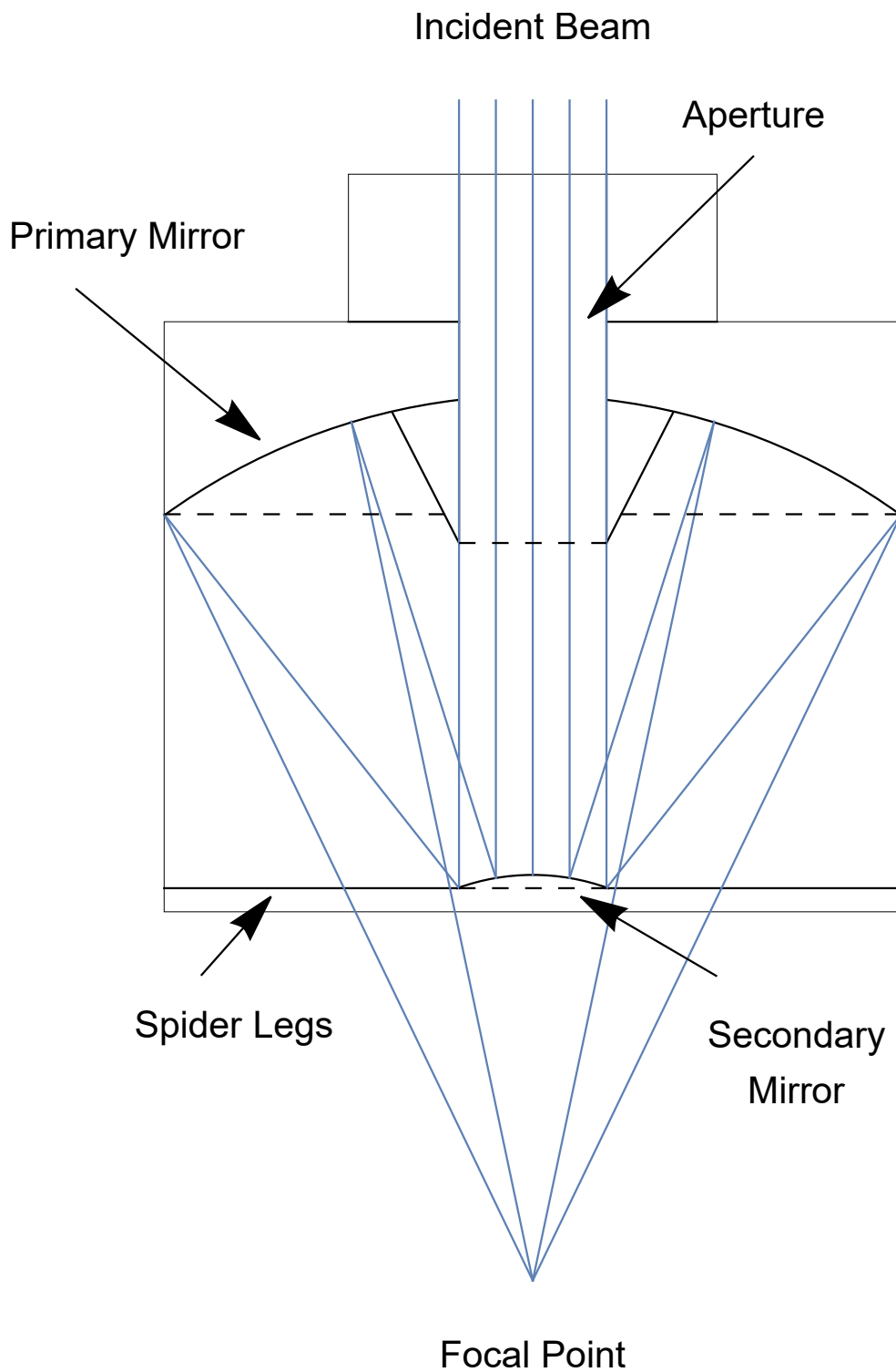


Figure 4.6: Schematic of a Schwarzschild reflective microscope objective with an infinite back focal length.

ing method requires calibration of the natural dose with several known laboratory doses. These data populate a dose response curve that is used to find the most recent burial or exposure date of the sample. There are several choices for a suitable radiation source, namely beta emitters – such as the conventional $^{90}\text{Sr}/^{90}\text{Y}$ – and X-ray emitters. X-ray sources emit two types of radiation: 1) characteristic X-rays at distinct energies that depend on the anode material and 2) a continuous bremsstrahlung-spectrum where the maximum energy depends on the high voltage applied between anode and cathode. A detailed description of X-ray source design, functionality and radiation properties can be found in Appendix C.

For the work presented here, an X-ray source has been chosen over a beta source due to four distinct advantages: 1) The X-ray source is much more uniform than the conventional $^{90}\text{Sr}/^{90}\text{Y}$ beta source (Spooner and Allsop 2000; Andersen, Bøtter-Jensen, and Murray 2003), 2) Due to the higher dose rate of X-ray tubes, the source to sample distance can be lengthened, providing a larger irradiation area up to 2.5 cm in diameter with comparable dose rates, 3) The X-ray tube has a variable potential and current, allowing for varied irradiation energies and dose rates, and 4) Shielding for radiation safety purposes is considerably easier for an X-ray source.

LuCIDD makes use of a 50 kV, hot-cathode X-ray source to provide these calibration doses and is capable of handling samples that are 2.54 x 5.08 cm – more than twice the size of samples the conventional reader can measure. With samples so large, it is important to characterize the dose rate, diameter, and uniformity of the irradiating beam to provide accurate irradiations along the entire sample surface. When applying long or multiple irradiations, the X-ray source will heat up and the dose rate could increase; as such, it is important to characterize the stability of the beam to determine any warm-up time required provide well-characterized doses. Another consideration is the uniformity of the photon flux and dose deposition. The conventional $^{90}\text{Sr}/^{90}\text{Y}$ beta source provides ionizing radiation with energies on the order of MeVs, and deposits its dose with a uni-

form flux within grains of sand that are $<125\text{ }\mu\text{m}$ thick. Depending on the energy of the X-ray beam, the photon flux could vary drastically at similar depths into solid samples. Hardening the energy of the X-ray beam is possible through the addition of an X-ray filter, reducing lower-energy, and thus less penetrating, X-rays and increasing the average energy deposited in the sample. Filtering an X-ray tube increases the overall measurement time by reducing the dose rate, while increasing dose deposition uniformity. A uniquely useful feature of an X-ray tube is its variable dose rate: for particularly sensitive samples, or if long irradiation times are desired, the input current of the X-ray tube may be decreased, lowering the X-ray output.

In 2003, Andersen, Bøtter-Jensen, and Murray performed characterizations on a 50 kV, 1 mA X-ray tube with a tungsten target by irradiating thermally-sensitized quartz sediments. They found that the X-ray source provided: 1) A dose rate that was linearly proportional to the operating current of the tube, 2) A large dynamic dose range from 2-2000 mGy/s at varying energies and currents, 3) short-term stability, and no required warmup time, and 4) a highly uniform irradiation area. Thomsen et. al. further refined the application of X-ray tubes to OSL dating in 2006 by characterizing two X-ray tubes. They verified earlier findings as well as developed their own: 1) The irradiation area is more uniform than that of the conventional $^{90}\text{Sr}/^{90}\text{Y}$ beta source, 2) Both X-ray tubes displayed highly linear relationships between current and dose rate, 3) the OSL signal and dose response curves are indistinguishable to those of the conventional beta source, and 4) Unlike the conventional source, the dose rate of the X-ray tubes is sample dependent.

During this dissertation, six characterizations have been made, verifying and adding to previous research in order for LuCIDD to make use of an X-ray source for spatially-resolved OSL dating: 1) The dose rate of the X-ray source has been determined for multiple samples due to the sample dependence found by Thomsen et. al., 2) The diameter and uniformity of the X-ray beam have been measured, 3) The short-term stability has been observed to devise what warmup time is required, 4) The energy spectrum of

the X-ray source has been measured, 5) The relationship between dose rate and sample depth has been explored, and 6) The linear relationship between dose rate and current has been explored. These characterizations are discussed in detail in chapter 7.

4.6 Summary

This chapter determined the challenges of cobble surface dating and how to apply non-conventional techniques to produce dose maps of sample surfaces. Laser stimulation is used to obtain the spatially-resolved signal required to produce dose map pixels. To prevent the measurement of luminescence from non-surface layers of a sample, a confocal microscope is used for signal discrimination and collection. To calibrate the collected signal to a known dose, an X-ray source is used. With the combination of these elements, an instrument may be built with all the components required to produce equivalent dose information. The next chapter will describe the assembly of the luminescence instrument, LuCIDD, that has been developed to produce dose maps of sample surfaces.

Chapter 5

Description of LuCIDD

The novel instrument LuCIDD (Luminescence instrument with Confocal and Imaging unit for Dating and Dosimetry) has been developed to acquire spatially resolved data on whole cobble surfaces by applying advanced stimulation and detection methods based on the working principles of a confocal microscope. A photo of the instrument is shown in figure 5.1. Spatial measurements are achieved by measuring luminescence from single points (pixels) on the surface of the cobble and combining these pixels to build a dose map of the cobble's surface. Prior to measurement, LuCIDD must scan the rough surface of the cobble with a non-stimulating red laser to determine the objective focal length for each measurement point. Once focused, a stimulating laser is applied to each of these pixels and the emitted luminescence is collected. The luminescence signals are calibrated against a known, laboratory dose to determine the equivalent dose (D_e) of each pixel and a dose map is formed. A stimulation source, signal collection device, radiation source, and heat supply are required to adapt and apply the conventional SAR procedure to LuCIDD.

LuCIDD was initially designed by Dr. Regina DeWitt and coworkers at Oklahoma State University using grant funds from the National Science Foundation (Major Research Instrumentation Program, grant numbers #0722552 and #1215060). In 2011, LuCIDD was transferred to East Carolina University, where Dr. Ken Jacobs constructed the basis

for the current setup of the instrument and Chris Bonnerup developed the control software. During this dissertation's research, LuCIDD's design was optimized, its components were integrated and characterized, dose response curves were created to test the feasibility of the SAR procedure with whole surfaces, and pixel sizes were experimentally determined.

LuCIDD integrates the four components required for SAR dating through its stimulation unit, confocal detection unit, X-ray unit, and sample stage/heating unit. Figure 5.2 provides a front-facing schematic of LuCIDD, including all four of its constituent units, as well as the remote computer control. The stimulation unit provides a variety of lasers with wavelengths capable of stimulating small points on the surfaces of different sample types. The confocal and detection unit allows the measurement and filtration of the signal, while rejecting luminescence that may be stimulated from below the surface of the cobble. The X-ray unit provides in-situ irradiation to calibrate the dose response of the sample. The sample stage and heating unit allows for the movement between the confocal and X-ray units and for the in-situ heat treatment of the sample. Finally, the PC control integrates the use of each unit to provide the automated measurements of whole cobble surfaces. This chapter describes each of LuCIDD's constituent units, its control software, and the final design optimizations. All part numbers used to construct LuCIDD may be found in appendix D.

5.1 Stimulation Unit

Rock surfaces are highly non-uniform and consist of a variety of minerals. In order to stimulate different minerals, LuCIDD's stimulation unit consists of three stimulating lasers of wavelengths 852 nm (IR; for feldspars), 532 nm (green; for dosimeters such as $\text{Al}_2\text{O}_3\text{:C}$), and 473 nm (blue; for quartz). Because red light has the lowest stimulation efficiency, a fourth, 637 nm (red) laser is added for focusing purposes. Each laser is expanded and collimated to homogenize its beam, and each laser's path can be inhibited

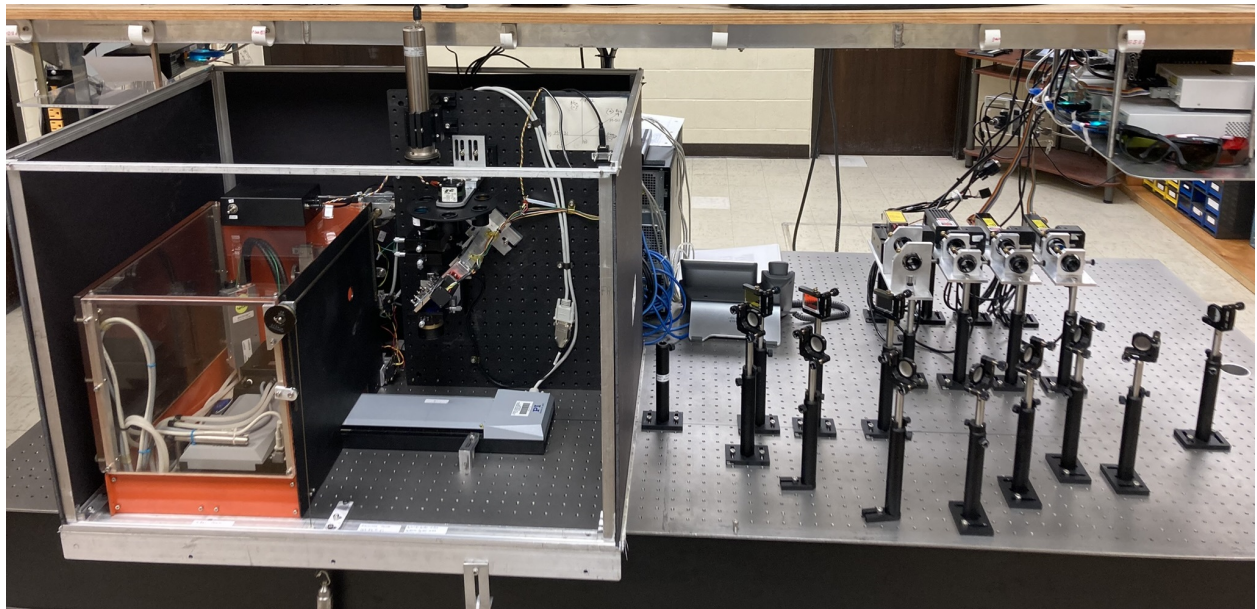


Figure 5.1: Assembled components of LuCIDD prior to adding the light-tight housing.

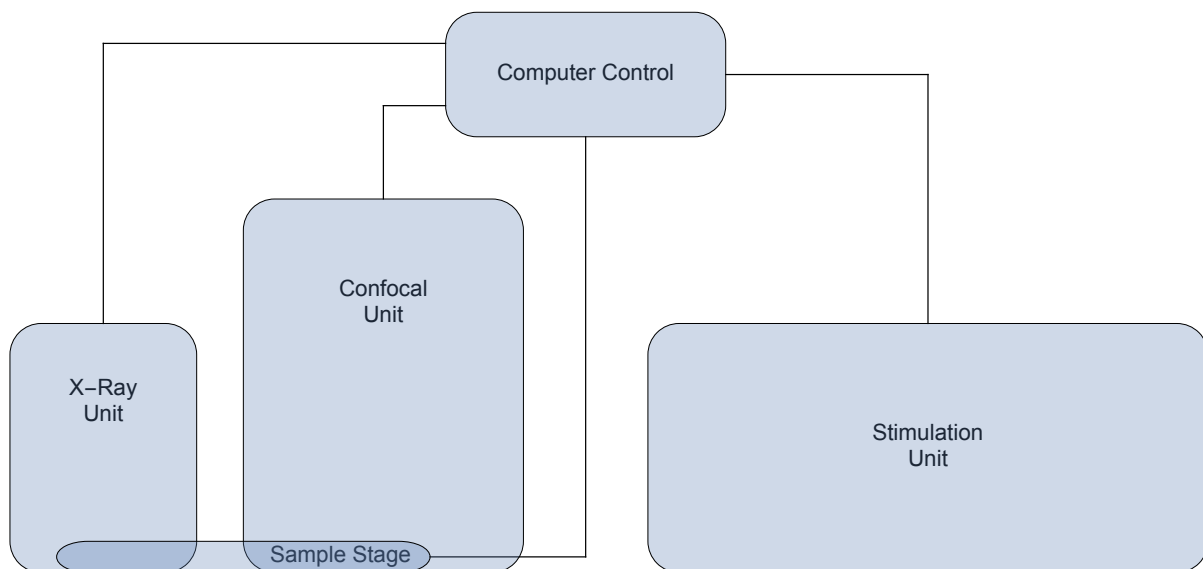


Figure 5.2: Front view schematic of LuCIDD. Each unit will be outlined in detail in the following sections.

by computer-controlled shutters that effectively “switch” the lasers on and off. Figure 5.3 is an image of LuCIDD’s stimulation unit.



Figure 5.3: Image of LuCIDD’s stimulation unit. All visible lasers were switched on for the photo, but only one will be used at a time during focusing and measurement. Only the high-power blue and green beams can be seen passing through the air.

The goal of the stimulation unit is to couple all four lasers and direct them into the confocal unit. This is accomplished by combining the beam paths of the lasers using a system of standard and dichroic mirrors. A schematic drawing of the four beam paths and a collection of dichroic mirrors is shown in figure 5.4. The lasers are coupled into a single beam path which is made through the unique properties of dichroic mirrors. Dichroic mirrors are specially coated to allow longer wavelengths to pass undeterred, while reflecting wavelengths equal to or shorter than the coating’s rating. For example, the “red” dichroic

mirror will pass wavelengths longer than 720 nm (i.e., passing the IR laser) while reflecting the red laser. The respective wavelength cutoffs for the green and blue dichroic mirrors are 570 nm and 500 nm, respectively (Fig. 5.5).

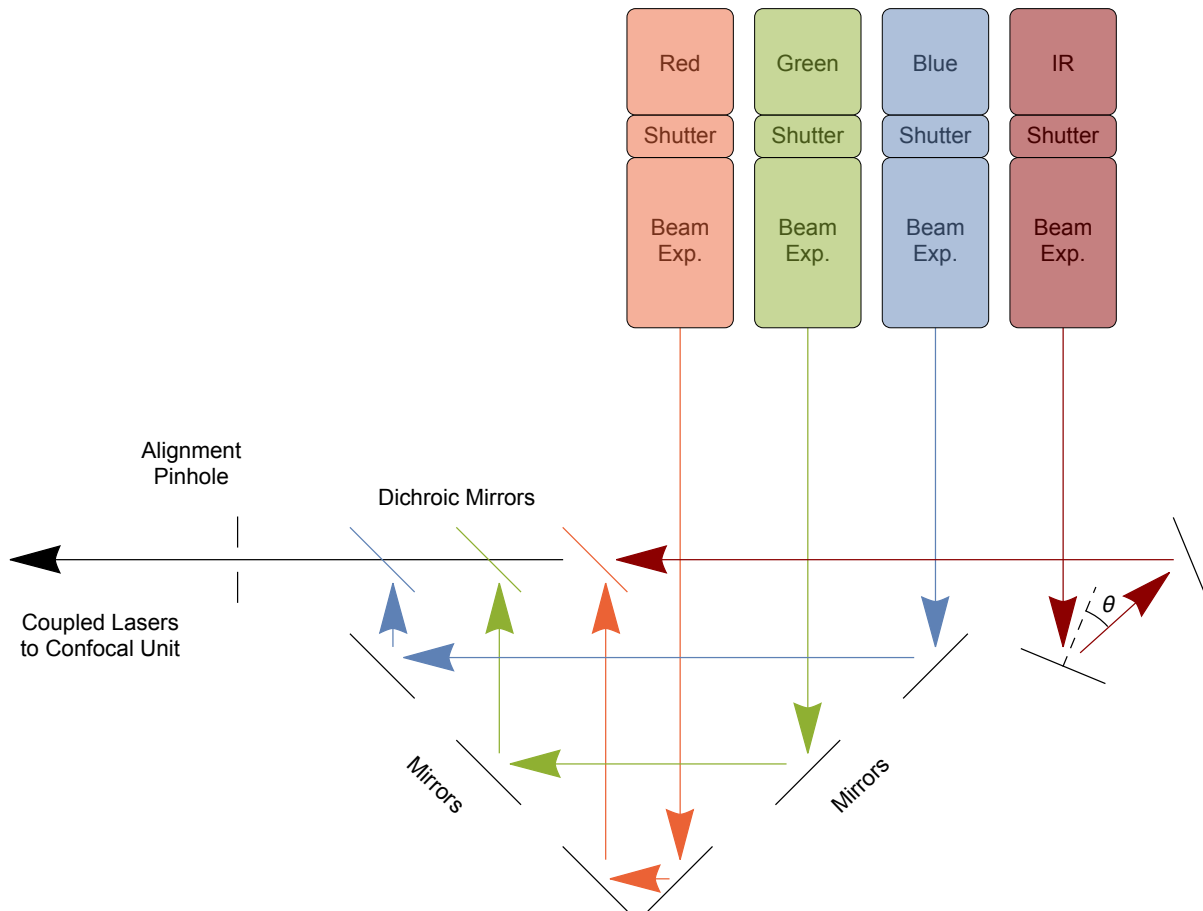


Figure 5.4: Top-down schematic drawing of LuCIDD's multi-laser system, which must be aligned to a single path using an assortment of standard and dichroic mirrors.

Once the lasers are aligned with the dichroic mirror assembly, they are considered coupled, and they pass from the dichroic assembly into the confocal and detection unit. To characterize the stimulation unit, the lasers were aligned, and three characteristics were determined for each stimulating laser: 1) focal spot size, 2) power density, and 3) penetration depth. These alignment and characterization procedures are detailed in chapter 6.

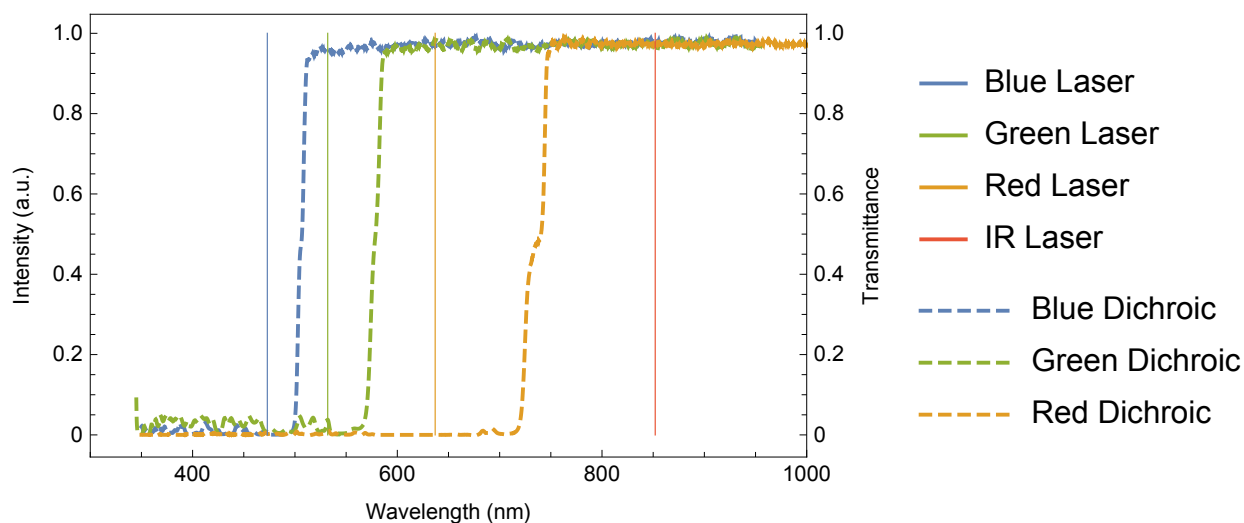


Figure 5.5: Wavelengths and transmittances of the stimulation unit's dichroic mirrors. The blue dichroic mirror is able to reflect the blue laser while passing the others. The green dichroic mirror will reflect both green and blue while passing the red and IR. The sequential placement of the dichroic mirrors is thus defined by their cut-off wavelength.

5.2 Confocal and Detection Unit

Recall from the previous chapter that lasers might bleach deeper into samples than sunlight will and thus stimulate luminescence from areas of the sample that have not been fully reset in nature. Measuring this luminescence results in the development of an incorrect dose response curve. The purpose of the confocal unit is to measure only signal originating from the surface of the sample, while blocking luminescence that is generated from below the sample's surface. To do this, a confocal microscope has been built into LuCIDD's confocal and detection unit. The confocal unit contains a second dichroic mirror assembly, an infinity-corrected reflective microscope objective, a confocal lens and pinhole system, and a photomultiplier tube (PMT) equipped with filters (Fig. 5.6).

When the selected laser passes from the stimulation unit into the confocal unit, it encounters the PC controlled dichroic mirror assembly, consisting of a beamsplitter and two dichroic mirrors. These two dichroic mirrors are specially coated to allow the stimulating laser to be reflected downward into the microscope objective, while allowing the lumi-

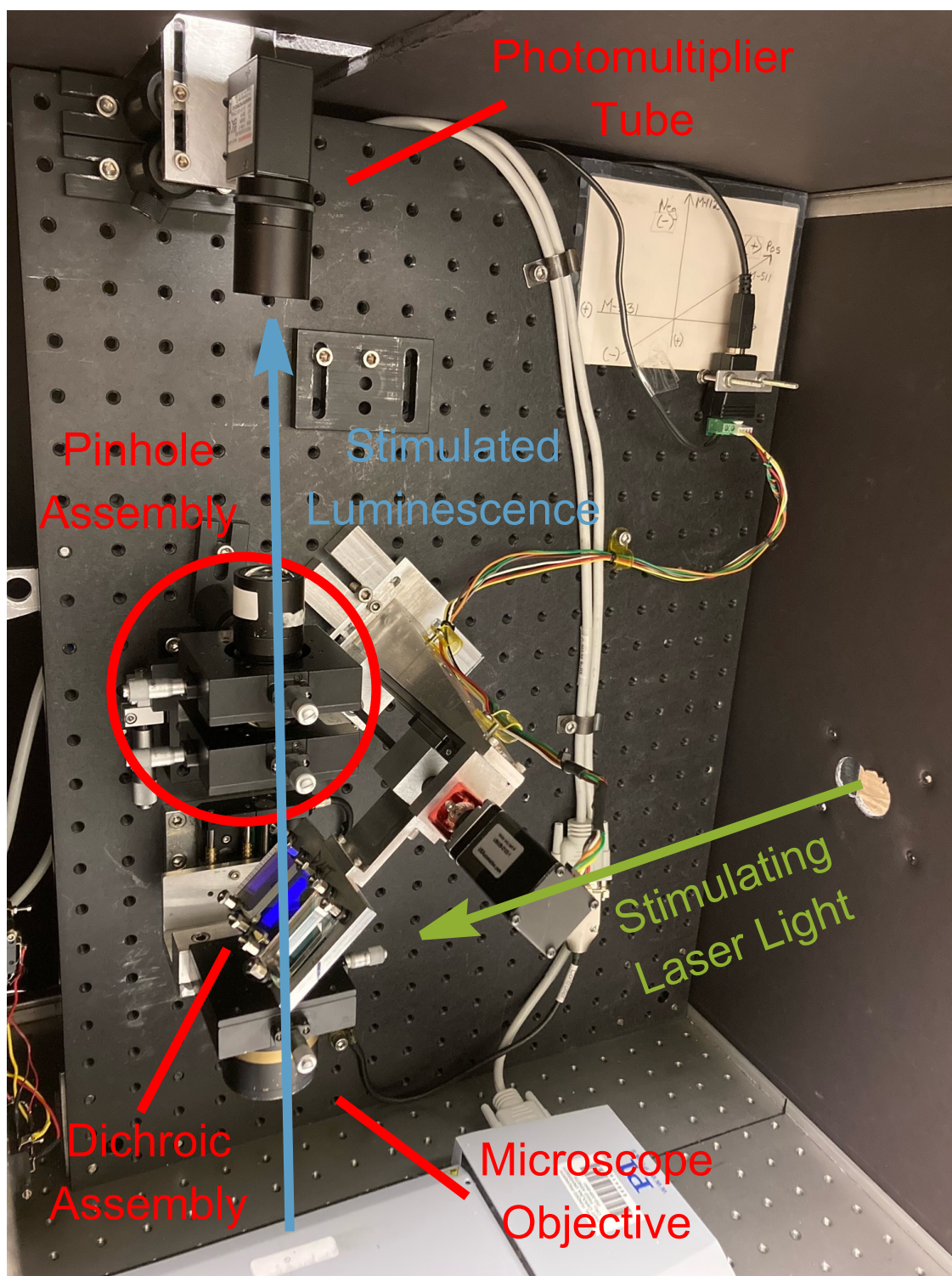


Figure 5.6: LuCIDD's confocal unit uses a dichroic mirror assembly to direct lasers into the reflective objective, focusing them onto the sample surface. The luminescence is passed through the objective, mirror assembly, and pinhole assembly to be captured by the PMT.

nescence to pass straight upward to the photomultiplier. The green (blue-pass) dichroic mirror reflects the IR and green lasers downwards and allows wavelengths from 390 nm to 500 nm to pass. The blue (UV-pass) mirror reflects the green and blue lasers and allows UV-wavelengths of 300 nm to 400 nm to pass. The mirror is selected depending on which sample type and stimulation wavelength will be used. Figure 5.7 shows the transmission wavelengths of the dichroic mirrors, and the emission wavelengths of the lasers. The Edmund Optics 50105-02 microscope objective has a magnification of 15x (NA 0.4), and is infinity corrected. This causes incoming parallel laser light to be directed into a narrow point in the focal point of the objective. A reflective objective was chosen because it will not suffer from chromatic aberrations, as opposed to refractive optics. This is required due to the wide range of wavelengths the objective will observe. The objective is mounted on a Physik Instrumente M-112 z-axis stage, allowing the focal length to be adjusted based on the surface height of the target pixel.

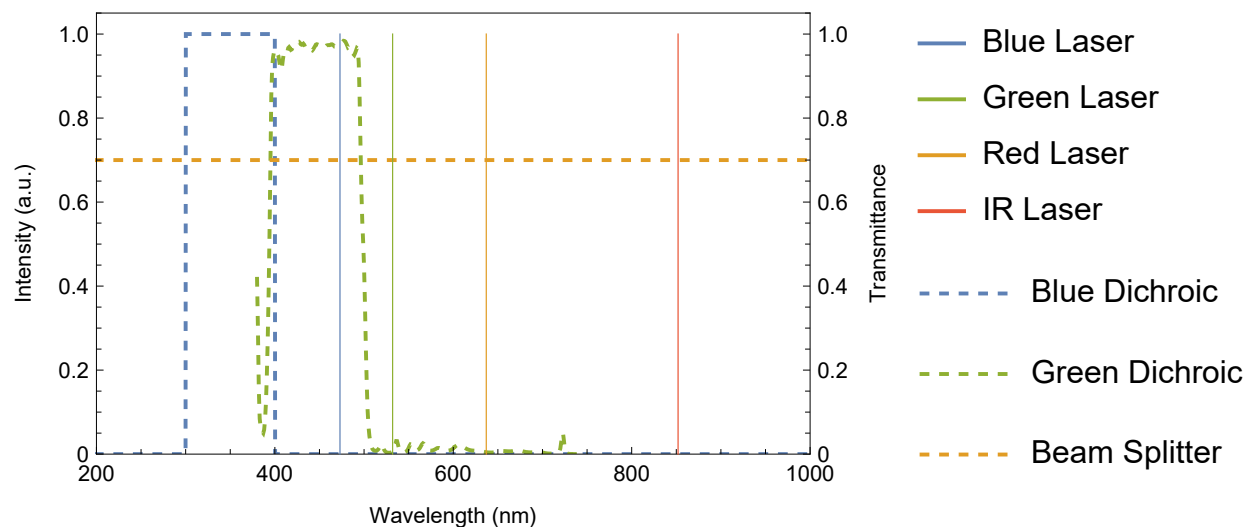


Figure 5.7: Transmission and emission wavelengths for LuCIDD's dichroic mirrors and lasers. The blue dichroic mirror is custom made, so the plot represents an estimation of the mirror's transmission wavelengths. A more accurate representation may be found in figure D.1.

Upon stimulating luminescence, both the laser light and the luminescence will travel

back through the objective. The laser light will be reflected away from the PMT by the dichroic mirror assembly while the luminescence passes through into the lens-pinhole-lens system. This system is designed to focus the luminescence through a 200 μm pinhole and into a Hamamatsu Photon Counting Head H11890 Series PMT. Much like the Risø luminescence reader, LuCIDD's PMT has an exchangeable filter that is used to block out any stray laser light and collect only luminescence as its signal. The filters used in this dissertation are described in section 8.1.

5.2.1 Focusing Method

To focus the objective, the red, non-stimulating laser is directed to the sample's surface using the dichroic mirror assembly's beamsplitter. At perfect focus, the beamsplitter allows the red laser to strike and reflect off the sample surface, through the detection pinhole, and into the PMT. With the laser switched on, the microscope objective traverses the z-axis stage and the PMT records the reflected laser's intensity. A neutral density filter with optical density 3.0 - i.e., an ND 3.0 filter - is used during focusing to prevent the PMT from being oversaturated or damaged.

While the objective is too far above the surface, reflected intensity is low. As the objective approaches a distance equal to the focal length, more laser light is transmitted through the pinhole until a maximum PMT count is observed. When the objective passes through the focal length, the PMT counts decrease. Figure 5.8 shows an example z-axis focusing scan made with the red laser. Considerations of LuCIDD's focusing method are examined in chapter 8.

5.3 X-Ray and Heating Units

Samples are mounted onto an Instec HCP322 hot plate that can be heated up to 300°C to provide the necessary temperature control during preheating and stimulation

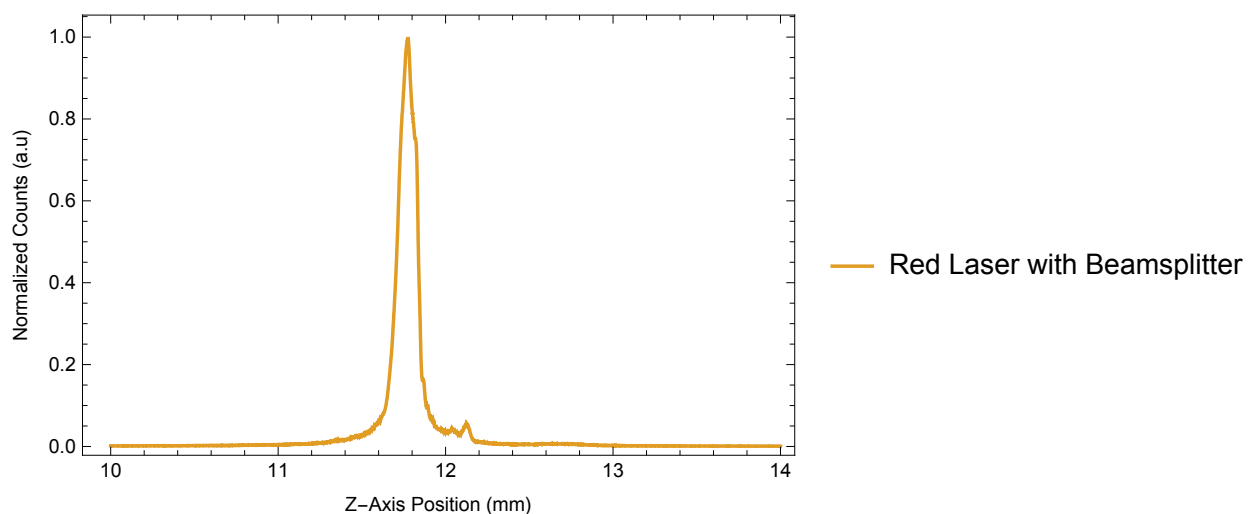


Figure 5.8: Reflected laser intensity versus microscope objective position. The position where the intensity is maximum is the position at which the laser beam on the surface has the smallest beam spot.

steps of the SAR procedure. As opposed to the Risø reader, the hot plate does not become oxidized at these temperatures, removing the need for bottled nitrogen gas. The hot plate is cooled using a PE-1 submersible water pump to provide room-temperature water through cooling channels within the hot plate.

The hot plate is mounted onto a remote controlled Physik Instrumente combined M-511 and M-531 x-y stage. This stage moves the sample in a horizontal plane beneath the microscope objective and enables scanning of the laser beam over the sample surface. The stage is also used to move the sample from the measurement position to the irradiation position under the X-ray source. The X-ray source is housed in an X-ray safe container made of lead-plexiglass which has a computer-controlled safety gate and interlock switches. The 50 kV, 200 μ A Magnum X-ray tube, is equipped with an 18 mm brass collimator and 50 μ m aluminum filter for irradiation (Fig. 5.9). The accurate production of a dose response curve majorly depends on the calibrating radiation source - the X-ray tube. To calibrate the X-ray source, the dose rate, beam diameter and uniformity, beam stability, energy spectrum, radiation penetration depth, and dose linearity with current were measured. These characterizations are described in detail in chapter 7.

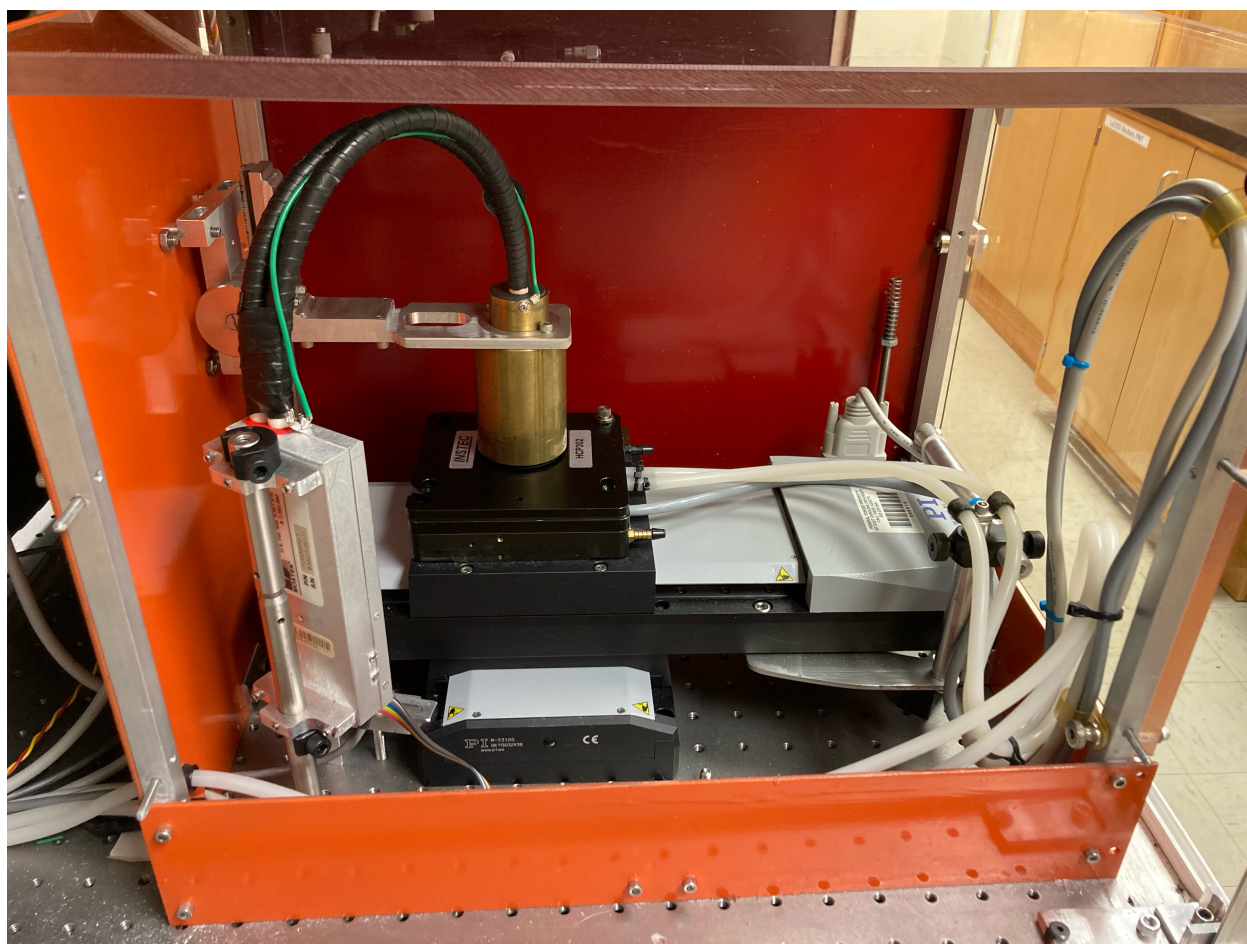


Figure 5.9: LuCIDD's built in X-ray housing allows for safe, quick irradiations without the sample leaving the instrument. The hot plate is heated electrically and cooled using room-temperature water.

5.4 Software and Control Features

Applying the full SAR procedure to a single aliquot takes hours and requires the timed application of a multitude of components. Operating each component in step with the SAR procedure is tedious and time consuming. A Lab View program has been developed by Chris Bonnerup at East Carolina University's Department of Physics to automate the SAR procedure using LuCIDD's hardware. The software integrates each required component and allows repeatable measurements with a single click. Figure 5.10 shows the LuCIDD Control Program's front panel.

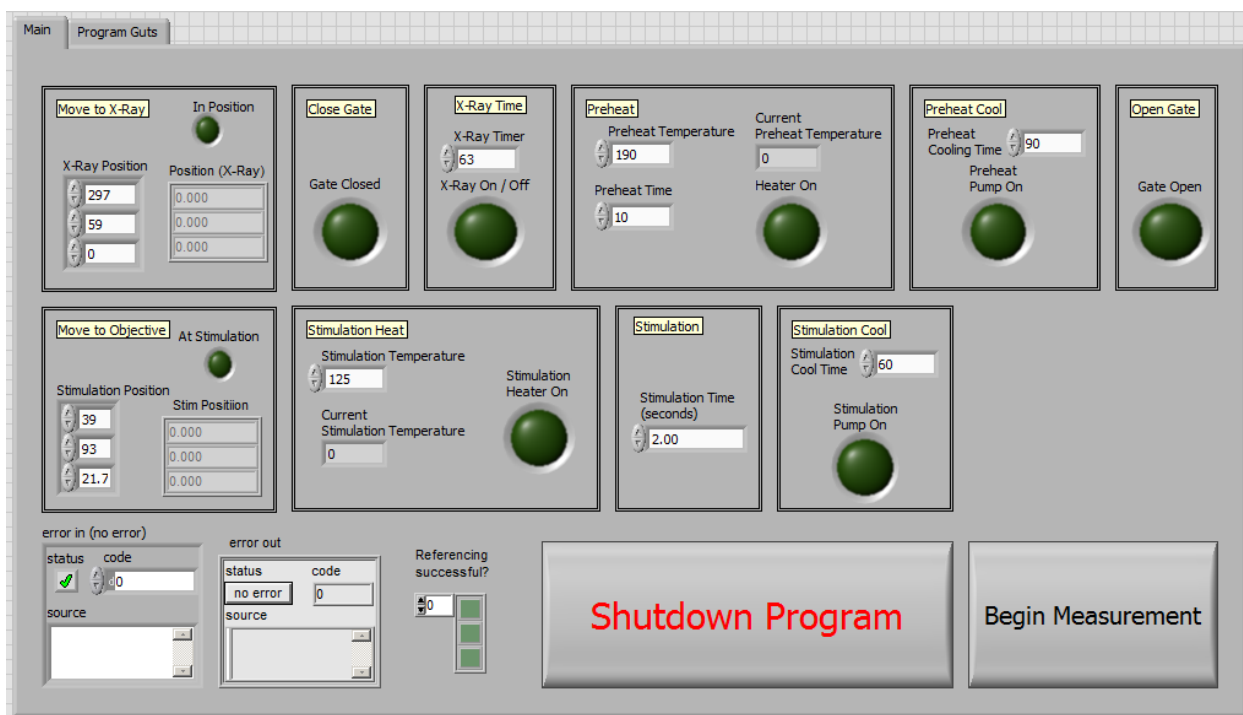


Figure 5.10: Screenshot of the front panel of the LuCIDD Control Program which integrates all the components required for performing the SAR procedure on a whole cobble surface.

The LuCIDD Control Program performs a single irradiation, preheat, and stimulation measurement, providing half of the required data to produce an SAR regeneration point (3.1). Executing the Control Program a second time allows the test dose, cutheat, and stimulation measurement to be performed, completing the regeneration point. The Con-

trol Program requires input for the irradiation position, regeneration (test dose) irradiation time, preheat (cutheat) temperature and hold time, post-preheat (cutheat) water pump cooling time, stimulation position, stimulation temperature, stimulation time, and post-stimulation water pump cooling time. When the program is first booted up, each LuCIDD component is initialized and referenced. When the initialization is complete, the “Referencing Successful?” lights will turn green to indicate that the program is ready to begin the measurement. Upon clicking “Begin Measurement,” the program will sequentially operate each of the steps a single time. Each step’s indicator lights turn green as the step is being performed, turning off again as the program moves on. When the final step is complete, the input values may be changed, and the program will be ready to run again. When all measurements are finished, LuCIDD’s components are safely de-initialized by clicking the “Shutdown Program” button and the individual components are able to be switched off. Any software errors are displayed in the error in and error out boxes. The data from each stimulation measurement is recorded by date and time in a folder on the user’s desktop.

It is important to note that, while the purpose of LuCIDD is to perform spatially resolved measurements, the LuCIDD Control Program currently only allows repeated measurements of a single point on the sample’s surface. To adapt the program for a second (nth) point, a second (nth) stimulation point must be specified and measured before the end of the Control Program’s cycle. Measurements to prove the ability of this instrument to perform spatially resolved measurements were performed manually, without the aid of automated software; however, the automated software did allow the creation of single-point dose response curves to prove LuCIDD’s ability to recover a known applied dose. These proof-of-concept measurements, as well as measurements of LuCIDD’s ability to provide reliable dose data are described in detail in chapter 8.

5.5 Optimizations From the Original Design

As mentioned above, the basic design and construction of the LuCIDD hardware was performed by Dr. Regina Dewitt and coworkers at Oklahoma State University as well as Dr. Ken Jacobs at East Carolina University. Many of these base design elements are still in use with small changes and the overall design of the instrument has stayed largely the same. However, many additions, changes, and tests led to the optimization of the construction of LuCIDD to improve ease of use, and simplicity of manufacturing. Some of the changes performed in the course of this dissertation are highlighted in the following paragraphs.

In the stimulation unit, the beam path has effectively remained the same, but updated laser, shutter, and beam expander mounts were applied. The laser shutter is now mounted prior to beam expansion, instead of after (Fig. 5.11). Before this improvement, the beam would be too large to fully fit through the shutter, leading to a reduction in power. Further, two removable alignment pinholes were added to the beamline to improve the reproducibility of the alignment procedures. The first pinhole is located between the stimulation unit and the confocal unit and is found at the end of the beamline in figure 5.4. The second pinhole is located beyond the X-ray source, where the gate and back panel of the X-ray housing may be removed to pass the laser unperturbed.

The changes to the confocal system began with the removal of a light collimator at the entrance of the unit. The original design of this collimator was a Keplerian beam expander (magnification=1) with a source pinhole at the convergence point between the two lenses. It was intended to “clean up” and collimate the stimulation source. Because each laser has a different wavelength, each laser would require individual collimators. This purpose is already served by the beam expanders in the stimulation unit. The removal of this source collimator also improved stimulation power by over 80%, allowing for shorter stimulation times and faster measurements. The second change made to the confocal system is

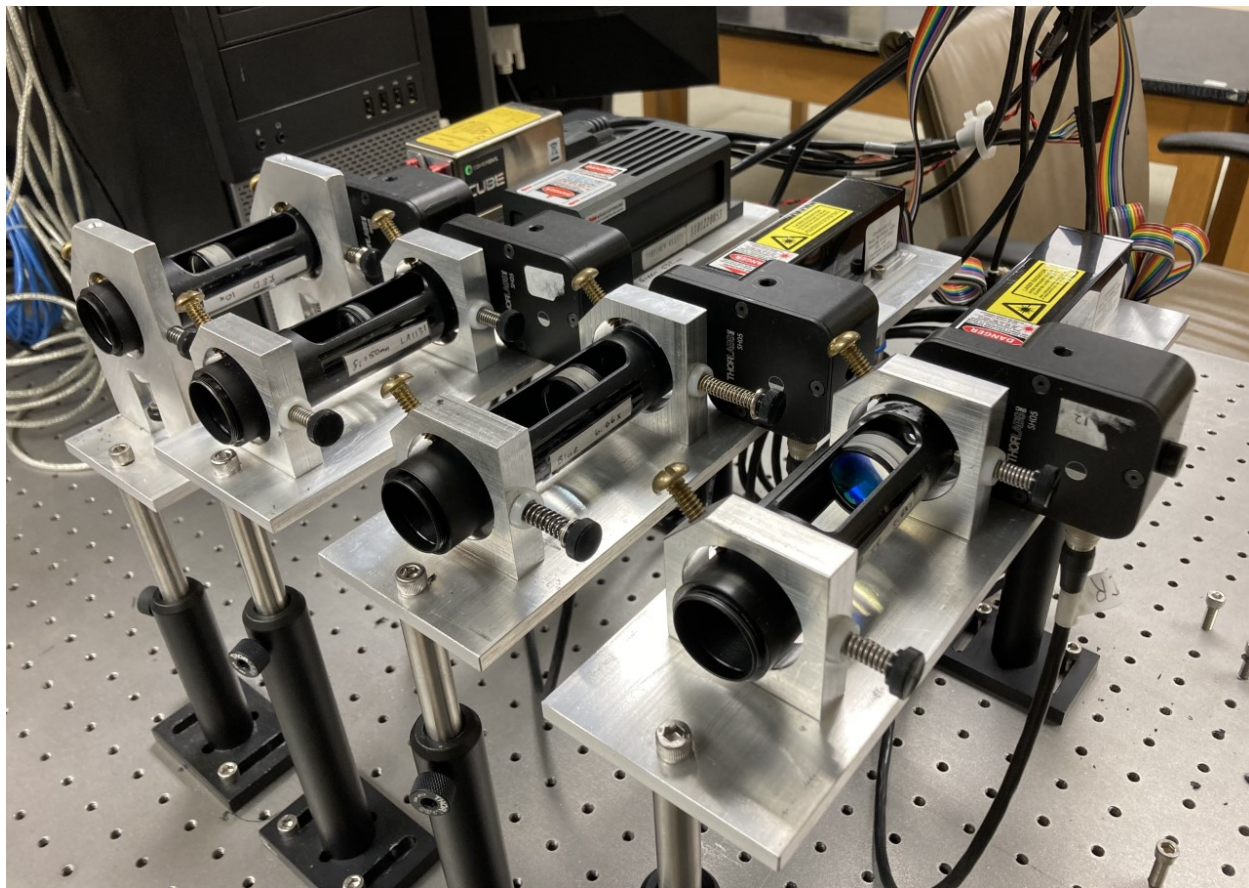


Figure 5.11: The updated laser mounts allow for adjustment of the beam expanders in two dimensions and add laser shutters between the lasers and the beam expanders.

an updated dichroic mirror assembly. Originally with four degrees of freedom, alignment of the dichroic assembly was near impossible. Removing the option for tilt and rotation and adjusting the axes of linear freedom to move in line with the mirrors themselves allows expedited, accurate, and easily reproducible alignment. The final change was to the objective mount with another four degrees of freedom. This mount was completely redesigned to remove two rotational degrees of freedom while still allowing the two linear degrees. The final design for the X-ray source is the same except for the addition of an 18 mm brass collimator, used to improve beam uniformity.

5.6 Summary

This chapter discussed the integration of the required components to perform luminescence measurements with spatial resolution. Lasers were coupled using multiple standard and dichroic mirrors and directed to a confocal microscope. The microscope has a dichroic mirror assembly which directs stimulation light downward to a sample, and allows luminescence signal to pass through to the detection pinhole and the PMT. The confocal microscope is focused on a sample's surface using a red non-stimulating laser. Calibrating irradiations are performed in a protected housing with an X-ray source. These components were all integrated electronically to be operated in tandem for the collection of spatially resolved equivalent dose information. Characterizations and tests of the lasers and X-ray components are found in chapter 6 and chapter 7, respectively. Final tests on LuCIDD are detailed in chapter 8.

Chapter 6

Stimulation Characterization

In chapter 4, it was determined that lasers could be used to develop dose maps of sample surfaces by stimulating and measuring luminescence from small “pixels” on the surface. The ability to use lasers for this application comes with two considerations: 1) The resolution of the instrument increases as pixel size decreases, which in turn increases the number of measurements and thus, time required to build a full dose map, and 2) Due to the high stimulation power, there is concern that lasers may penetrate to a depth in the sample beyond what the sample would experience in nature by the sun. This chapter describes the characterization of LuCIDD’s stimulation unit beginning with the instrumentation required to test the lasers. The procedure for aligning the four lasers and combining their separate beam paths is discussed in the second section. Finally, the lasers’ properties are summarized, and their penetration depths are quantified.

6.1 Instrumentation

LuCIDD is equipped with four lasers that were aligned and characterized for focal spot size, power density, and penetration depth using a Thorlabs BC106-VIS CCD laser camera and an Ophir Nova II power meter with Ophir PD300-3W power sensor. Data

collected with these devices were processed and visualized using Wolfram Mathematica.

6.1.1 Laser Camera

A Thorlabs BC106-VIS CCD laser camera was used to align and measure the focal spot size of each of the four lasers (Fig. 6.1). The camera was equipped with an ND 40 neutral density filter to prevent the lasers from saturating the CCD chip. Even so, the lasers were run at power levels lower than their maximum output to prevent damage to the laser camera. Each pixel of the CCD chip is $6.45 \times 6.45 \mu\text{m}^2$, and the lower measurement limit of the camera is 10 pixels wide, limiting a minimum acceptable measurement of beam width to $64.5 \mu\text{m}$ in diameter.

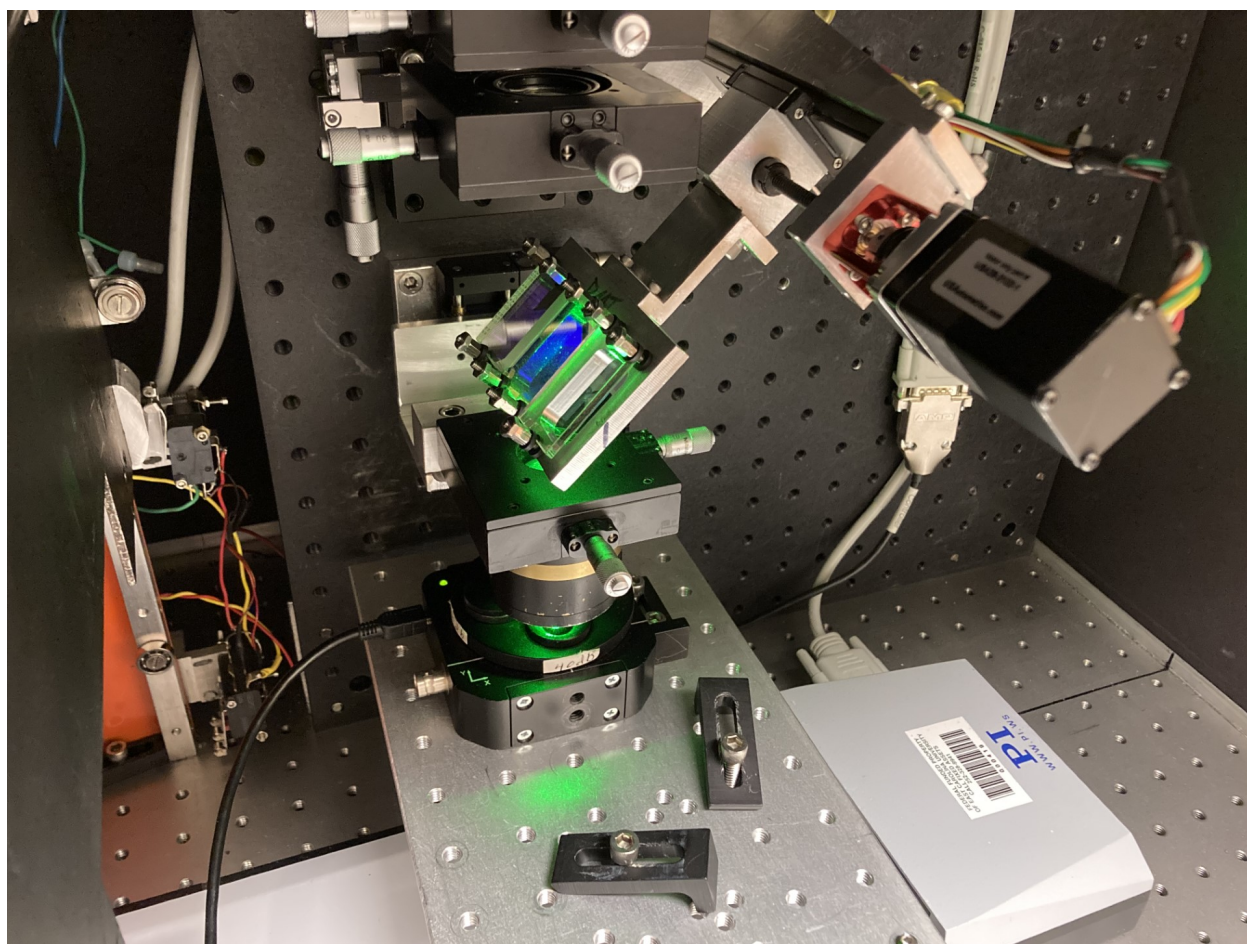


Figure 6.1: Image of the laser camera experimental setup. Lasers are focused onto the camera's CCD chip to measure their beam diameters.

The BC106-VIS CCD laser camera sends live measurements directly to its mother program: Thorlabs Beam (Fig. 6.2). This program requires an ambient light correction as well as user input of the ND absorbance of the equipped filter and the wavelength of the selected laser. Exposure time and gain may be set manually, but automatic values were used for the measurements described in this chapter. To measure the focal spot sizes, measurements of the 4-sigma beam width were taken by the Thorlabs Beam software and averaged using Mathematica. The 4-sigma beam width $D_{4\sigma}$ as measured in the $x - y$ plane is the International Organization for Standardization's standard for measuring the diameter of a circularly symmetric laser beam (*Optics and photonics – Lasers and laser-related equipment – Vocabulary and symbols* 2018):

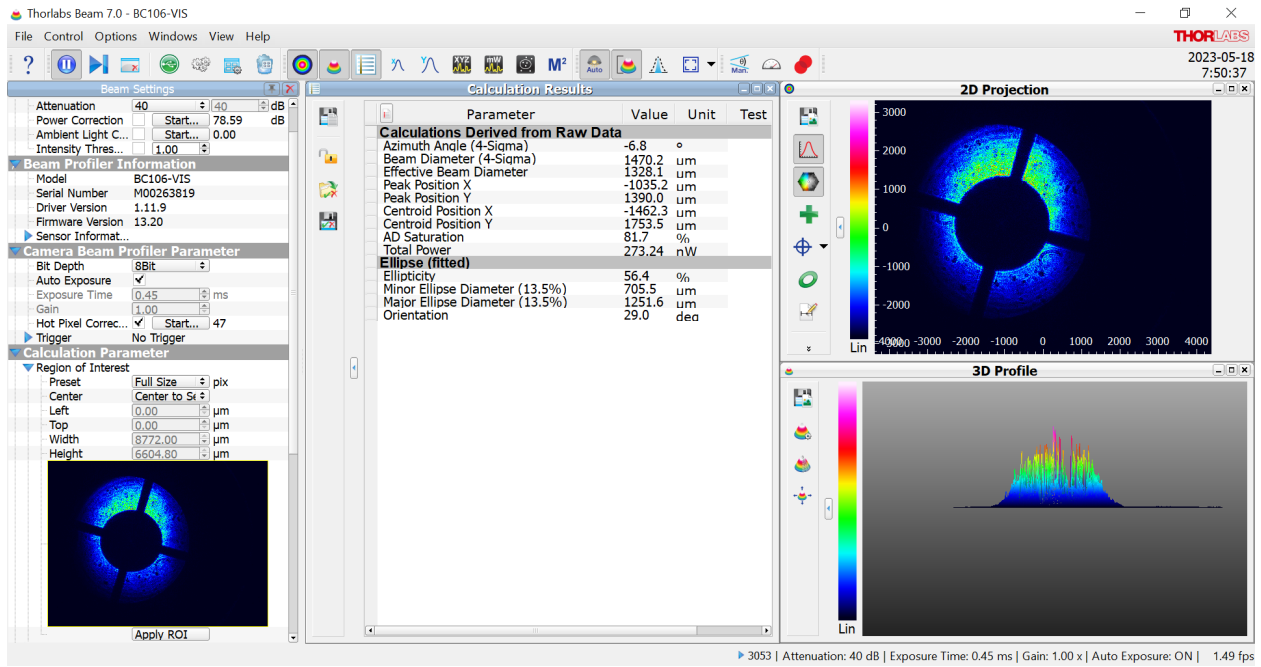


Figure 6.2: The laser camera's software provides live output of a variety of measurements, as well as an image of the beam. This output is of a laser before reaching its focal point. The obscuration due to the primary mirror and spider legs of the reflective objective can be clearly seen.

$$D_{4\sigma} = 2\sqrt{2} \sqrt{\frac{\sum_{x,y} [p(x,y)((x - \bar{x})^2 + (y - \bar{y})^2)]}{\sum_{x,y} [p(x,y)]}} \quad (6.1)$$

Where $p(x, y)$ is the intensity measured at the pixel location (x, y) of the CCD chip and $\bar{x}$ and $\bar{y}$ are the pixel coordinates for the centroid of the beam determined by the following equations:

$$\begin{aligned}\bar{x} &= \frac{\sum_{x,y} [p(x, y)x]}{\sum_{x,y} [p(x, y)]} \\ \bar{y} &= \frac{\sum_{x,y} [p(x, y)y]}{\sum_{x,y} [p(x, y)]}\end{aligned}\tag{6.2}$$

6.1.2 Laser Power Sensor

An Ophir Nova II power meter was used in conjunction with an Ophir PD300-3W power sensor to determine the power density and penetration depth of the stimulating lasers using thin sectioned rock slices. Figure 6.3 shows an image of the green laser incident on the power sensor as well as the power meter output. The laser sensor has an attached filter which allows measurements of laser power from 2 μ W to 3 W and requires input of the selected laser's wavelength. The power of each laser was measured at the focal point of the microscope objective to ensure that the measured power is the same as that used for stimulating a sample spot. The power measurement was taken as a five-minute time average and was divided by the total area of the laser's focal spot size to determine the power density.

6.2 Alignment

Each laser was aligned using the laser camera and multiple alignment pinholes. The alignment of the lasers was divided into the major sections of the optical path: 1) Each laser, mount, and beam expander, 2) The stimulation unit, 3) The vertical area below the confocal dichroic mirror assembly, and 4) The vertical area above the confocal dichroic mirror assembly.

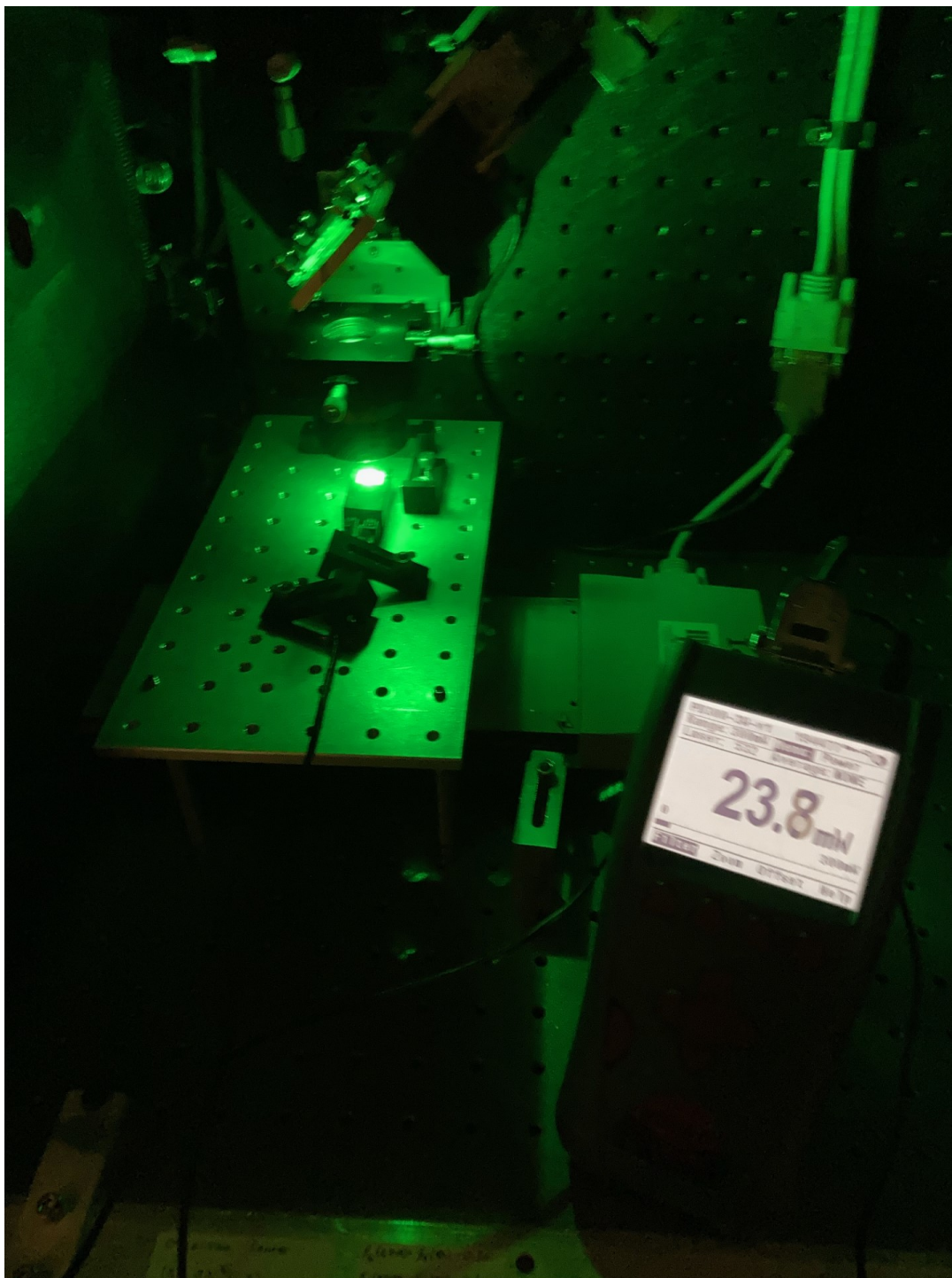


Figure 6.3: The power sensor was placed beneath the microscope objective to measure each laser's integrated power.

1) Each laser and beam expander was attached to its mount and the laser beam was directed straight ahead toward the wall of the laboratory. The beam expander components for each laser were chosen to provide a beam that would fully fill the objective's aperture (9.728 mm). Over filling the aperture of the objective results in a total loss of the overfilled portion of the beam; underfilling the aperture causes a decrease in power due to the obscuration spot of the reflective objective. The beam expander's lenses (Fig. 4.3) were adjusted so the measured width of the beam was the same at any location on the laser's path, creating a collimated and parallel beam.

2) Each individual laser was reflected by two mirrors (Fig. 5.4) before impinging upon the stimulation dichroic mirror assembly. Upon striking the dichroic mirror assembly, the laser paths were combined into a single path and continued into the confocal unit. To proceed with alignment, removal of the confocal dichroic mirror assembly, X-ray source, and one wall of the X-ray housing was required. Each laser was verified to be aligned horizontally with the optical table using two alignment pinholes of equal height. The first pinhole, which is shown in figure 5.4, was placed at the very end of the stimulation unit beam path. The second pinhole was placed beyond the X-ray housing at the edge of the optical table to provide the longest path possible for alignment. The lasers were adjusted until they passed through both alignment pinholes and hit the same point on the far wall of the laboratory.

3) The removed components were put back in place, while the reflective objective was removed and replaced with a set of two alignment pinholes using a lens tube. Each dichroic mirror was aligned such that its respective laser would travel vertically down through the pinholes to strike the sample area.

4) The alignment pinholes were removed from the laser path and a planar mirror was placed onto the sample area. The planar mirror reflected each laser back (i.e., upwards) through the confocal dichroic mirror assembly and into the first lens of the confocal pinhole assembly. The confocal pinhole was placed into the laser's path and the first lens was

adjusted until a sharp image was observed on the far side of the pinhole. The second lens of the assembly was added to focus the light into the photomultiplier tube (PMT). Note that the PMT had a light-proof cap on during the alignment process to prevent it from becoming damaged by the high power lasers. Once the beam path was fully aligned, the reflective objective was put in place and the instrument was ready to proceed with measurements.

Alignment of the lasers was a laborious and time-consuming process due to the number of components, the degrees of freedom and movement for mirrors and mounts, and the requirement that all lasers must be coupled to strike the same spot on the sample surface. The alignment procedure outlined in this section was the culmination of weeks of alignment tests, beam path optimization, and optical mount optimizations. Misaligned lasers could result in a decrease of stimulating power, enlarged focal spot diameter, and stimulation of incorrect areas of the sample's surface.

6.3 Focal Spot Size

The focal spot size of each of the four lasers was determined using the CCD laser camera. The laser camera was placed underneath the microscope objective in the same location a sample would be placed. The z-axis of the objective was adjusted until the minimum beam diameter was observed (Fig. 6.4). Each beam's diameter was measured 100 times and averaged, providing the values found in table 6.1. Note that each of these measurements results in a spot that is smaller than 10 pixels of the laser camera – the lower limit of accuracy provided by the BC106-VIS camera. Thus, calculations requiring beam diameter will be performed with both the measured values, as well as the lower measurement limit of $10 \times 6.45 \mu\text{m} = 64.5 \mu\text{m}$.

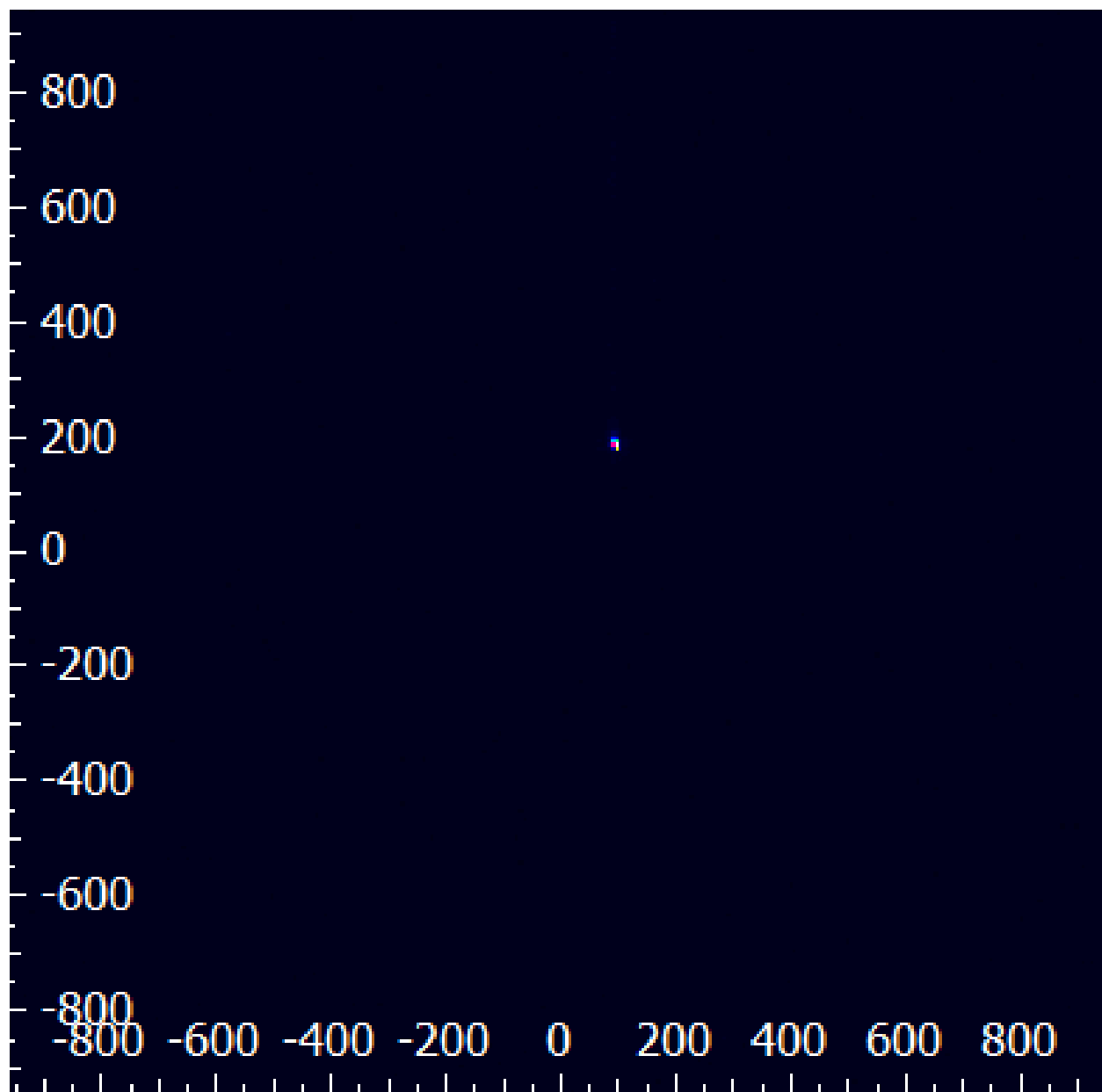


Figure 6.4: The focal spot of the green laser on the camera output. The beam appears to be only 2-3 pixels wide.

Table 6.1: Laser Beam Focal Spot Diameters. Listed are the lasers and the respective dichroic mirrors that were used to guide the lasers into the microscope objective.

Laser	Dichroic Mirror	4σ Width (μm)
Blue	Blue	21.514 ± 0.022
Green	Green	21.414 ± 0.027
IR	Green	21.19 ± 0.06
Red	Beamsplitter	41.100 ± 0.021

6.4 Power Density

The power density of a laser's focal spot is the energy imparted by the laser on the incident area per second, directly related to the photon flux. Larger power densities will allow more photons to stimulate luminescence from a sample. In smaller areas, this equates to shorter stimulation times for the same output signal. However, having gaussian profiles, there is a probability for the photons to be deposited in areas outside of the focal spot of the laser, bleaching those areas. A larger power density will cause larger photon fluxes that will stimulate these external areas more often.

For each laser, except the red non-stimulating laser, the power can be adjusted with a control knob. A laser power sensor was used to measure the stimulation power of each laser in the focal point of the objective. The power was divided by the area of the stimulating laser's focal spot - using the measured values, as well as the value $r = 32.25\mu\text{m}$ from the previous section - to determine the power density. Figure 6.5 shows the measured power density for the three variable lasers (green, blue, and IR) in dependence of the controller output setting. It can be seen that the green laser has a reduction in power density when the controller is set between 73% and 80%. This was consistently reproducible and is thus far unexplained. Each laser's maximum power density is shown in table 6.2.

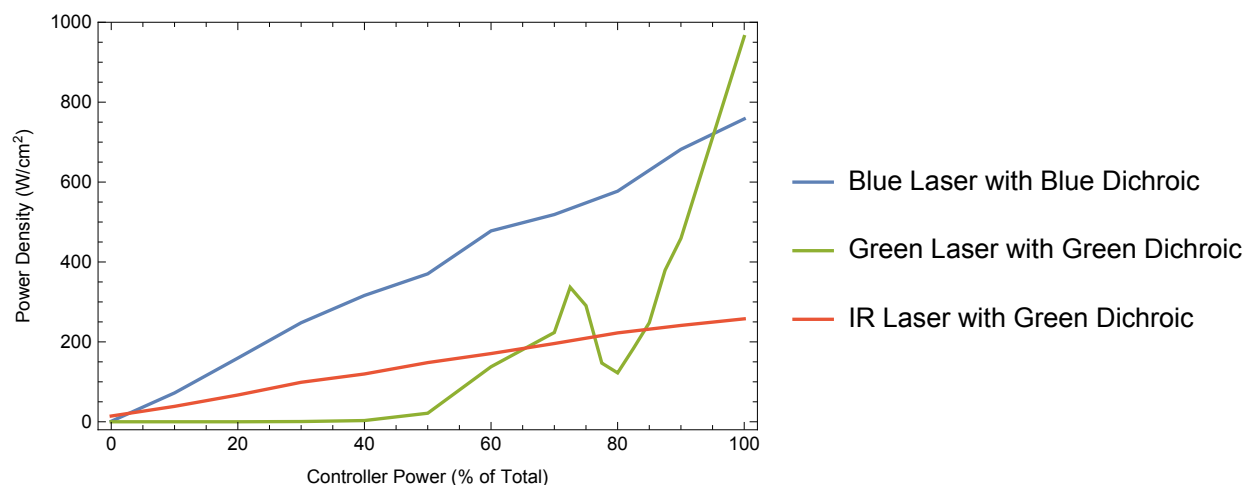


Figure 6.5: The power density of the laser focal spot relative to the laser controller's output setting. The green laser has the highest power density of the three laser and mirror combinations but has an unexplained dip in power output at certain controller settings.

Table 6.2: Power densities for varying laser and dichroic mirror combinations. The theoretical power densities are also displayed here, calculated using the measured beam radii.

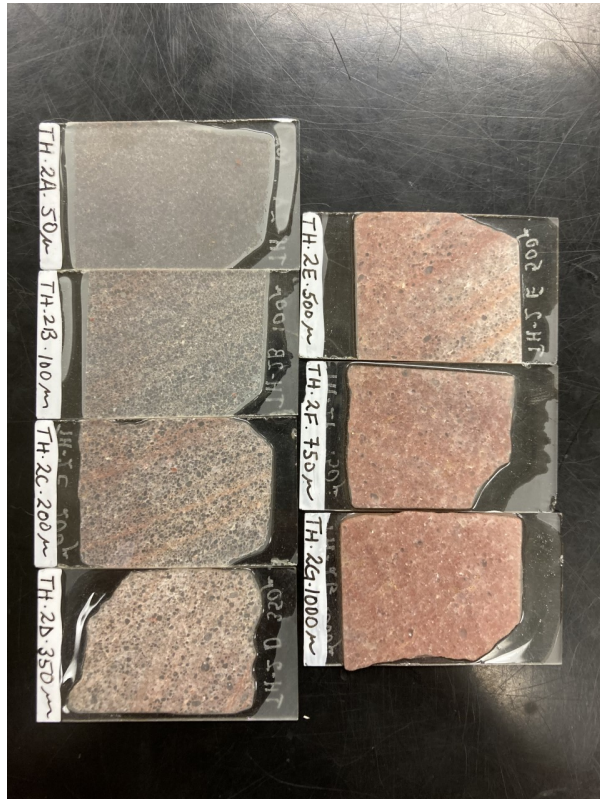
Laser	Dichroic Mirror	Measured Power (mW)	Power Density at $r = 32.25 \mu\text{m}$ (W/cm^2)	Power Density at measured r (W/cm^2)
Blue	Blue	24.77	758	6814 ± 14
Green	Green	31.50	964	8746 ± 22
IR	Green	8.42	258	2388 ± 14
Red	Beamsplitter	0.4	1.2	3.0150 ± 0.0031

6.5 Penetration Depth

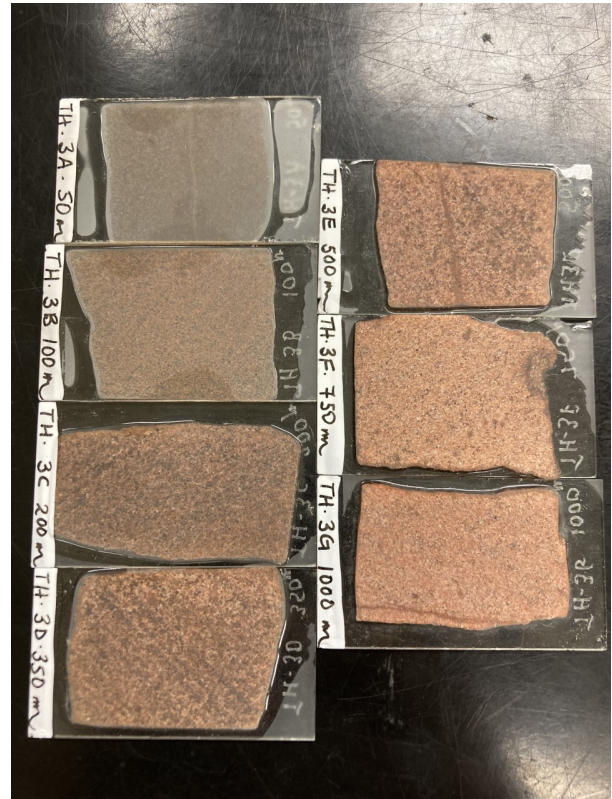
In chapter 4 it was determined that sunlight could bleach OSL traps as deep as 1 mm in only an hour of exposure, and that OSL traps will bleach faster than IRSL traps will (Simms et al. 2011; Fuhrmann et al. 2022). While not analogous to bleaching depth, the penetration depths of the lasers provide a time-independent approximation for the phenomenon of bleaching. LuCIDD's purpose is to perform spatially resolved measurements through the application of a confocal microscope, preventing sub-surface OSL from reaching the PMT. Thus, signal that originates beyond the 1 mm depth, as is used in conventional rock measurements, is removed. Even though LuCIDD has this safeguard, it is important to understand the depth at which the lasers can penetrate into and stimulate OSL from rock surfaces.

To determine the penetration depth of the lasers, quartzite and sandstone thin sections were purchased from Vancouver Petrographics Ltd (Fig. 6.6). The thin sections consisted of rock that had been glued onto the surface of a cover slip, ground flat to the desired thickness, and then polished. The thin sections used in this research had thicknesses of 50-1000 μm and were not homogenous, having variations of grain size, mineral composition, and opacity. Prior to measurements, the slides were cleaned of smudges and fingerprints using denatured alcohol and an optical cleaning wipe.

The laser power sensor was used to determine each stimulating laser's ability to penetrate into and pass through quartzite and sandstone. Thin sections of thicknesses from 50 μm to 1000 μm were placed in front of the power meter for measurement. The laser was focused onto selected transparent and opaque spots on the surface of the thin sections. These were considered to be representative of the maximum and minimum transmission, and thus penetration, into the rock. The transmitted power was measured for the various grains and thin section thicknesses. The thin sections were then replaced with an empty coverslip and the power was again recorded. The thin section power was divided by the



(a) Quartzite Thin Sections



(b) Sandstone Thin Sections

Figure 6.6: Quartzite and sandstone thin sections used for quantifying penetration of the laser into the sample. The thin sections are not homogenous and individual grains of differing opacity are apparent.

cover slip power to determine the percent of transmitted power at each depth. Theoretically, transmitted laser intensity I in a material with a constant absorption coefficient a will decrease with depth z according to the Beer-Lambert law:

$$I = I_0 e^{-az} \quad (6.3)$$

The optical penetration depth is defined as the value of z where $I = I_0/e$, i.e., $z = 1/a$. This is only true for a well-behaved absorption coefficient, which usually varies based on sample temperature and the wavelength of the applied light. Figure 6.7 shows the wavelength dependence on optical penetration depth of single-crystal silicon (Green and Keevers 1995).

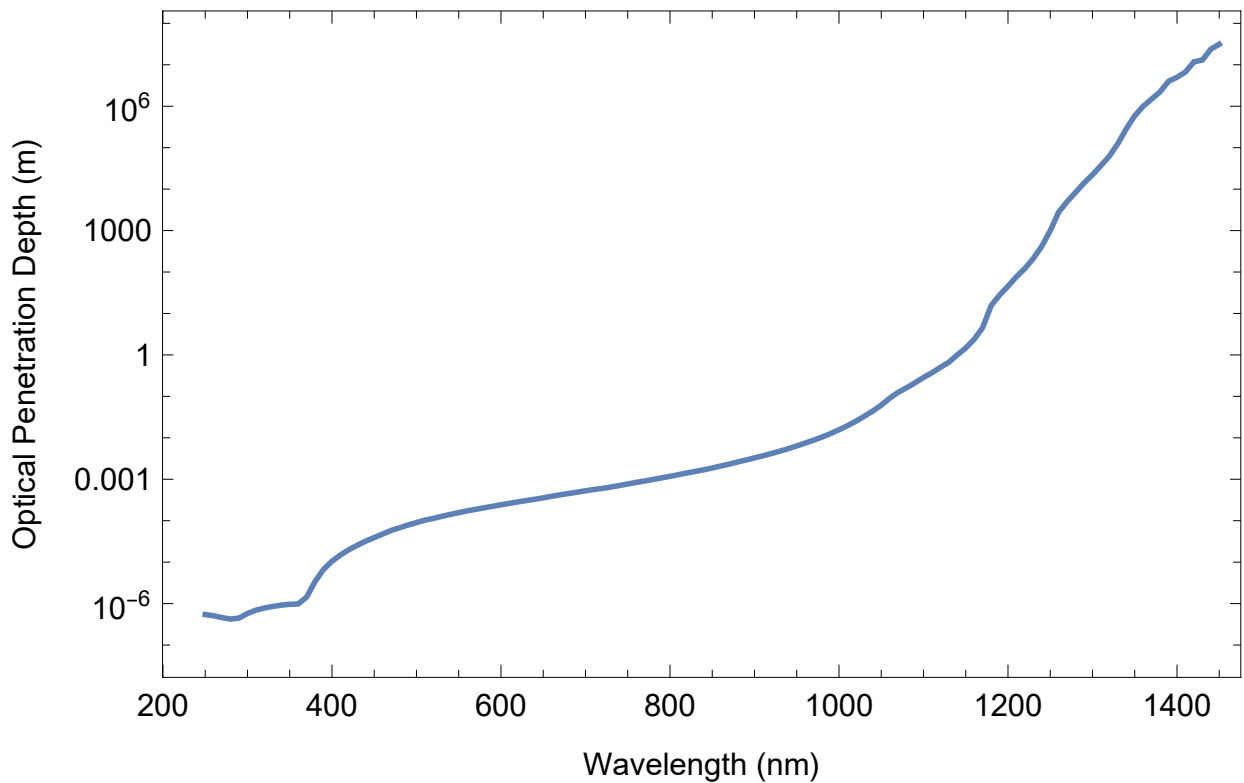


Figure 6.7: The optical penetration depth of silicon increases dramatically with higher wavelengths of light, with blue 470 nm light penetrating 58.1 μm into the sample, and IR 870 nm light penetrating 2.31 mm.

Figure 6.8 shows the transmitted power of multiple laser and mirror combinations for

sandstone. Curves were fit with the form of the Beer-Lambert law, but only measurements made on the opaque grains followed the fit. Table 6.3 shows the penetration depth for each laser and mirror combination, material, and opacity. The opacity of the material greatly affected the penetration depth, decreasing the values to one tenth of those observed in the translucent areas. In all cases but one, the penetration depth in quartzite was greater than the penetration depth in sandstone and all penetration depths are less than the 1 mm threshold that is used in conventional dating of rock surfaces (Sec. 4.1).

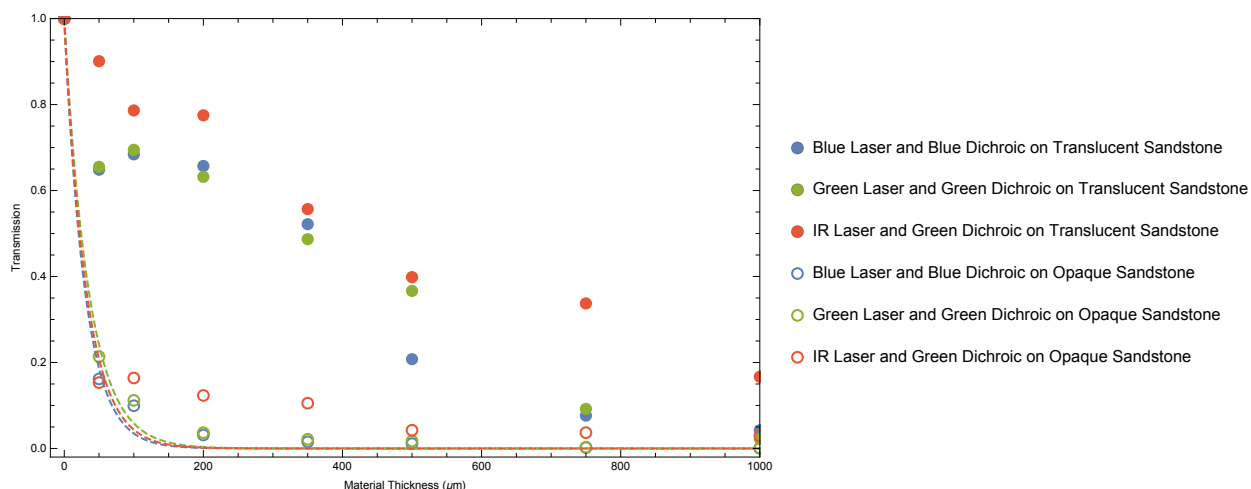


Figure 6.8: Each laser penetrates farther into translucent areas of the thin sections than the opaque areas. The fits for the opaque areas roughly follow the Beer-Lambert law for the attenuation of light into a medium. All penetration depths are less than 1 mm within the surface of the sample.

6.6 Discussion

The diameters for the focused stimulation lasers were all found to be below the minimum threshold for the laser camera: $<64.5 \mu\text{m}$. This minimum threshold is just over 50% of the diameter obtained by the single-grain measurement device ($120 \mu\text{m}$) and just over 25% of the beam diameter that others have used for spatially-resolved dating (Duller et al. 1999; Bailiff and Mikhailik 2003). While the $64.5 \mu\text{m}$ theoretical limit of the beam diameter is already small compared to values quoted in other publications, the true focal diameters

Table 6.3: Laser Penetration Depth in Different Materials

Laser	Dichroic Mirror	Material	Opacity	Penetration Depth (μm)
Blue	Blue	Quartzite	Translucent	682
Green	Blue	Quartzite	Translucent	764
Green	Green	Quartzite	Translucent	706
IR	Green	Quartzite	Translucent	961
Blue	Blue	Sandstone	Translucent	375
Green	Blue	Sandstone	Translucent	456
Green	Green	Sandstone	Translucent	406
IR	Green	Sandstone	Translucent	607
Blue	Blue	Quartzite	Opaque	54
Green	Blue	Quartzite	Opaque	41
Green	Green	Quartzite	Opaque	49
IR	Green	Quartzite	Opaque	55
Blue	Blue	Sandstone	Opaque	30
Green	Blue	Sandstone	Opaque	53
Green	Green	Sandstone	Opaque	35
IR	Green	Sandstone	Opaque	32

of the lasers are actually much smaller. Further decreases in the beam diameter would equate to an increased calculated power density at the focal length of the confocal microscope proportional to $1/r^2$. A smaller focal spot size would ideally result in a smaller pixel size and greater dose map resolution, and these characterizations will be explored in more depth in chapter 8.

The power density for each laser was characterized at different controller power settings, providing values on the order of $10^2 - 10^3 \text{ W/cm}^2$, as compared to the laser power density of about 50 W/cm^2 for the Risø single-grain attachment (Duller et al. 1999). The power densities are thought to be even higher if more precise measurements of the beam diameters are made. At this time, the impact of power density on the true bleaching radius of each laser is unclear.

Each laser's penetration depth was determined in sandstone and quartzite rock thin-sections for both opaque and translucent grains. It is important to note that the data did

not closely follow the fit curves used to determine these penetration depths. The depth where the laser's power became $1/e \approx 36.8\%$ of the incident power for each laser was less than 1 mm for the translucent grains and at most 55 μm for the opaque grains. These depths are less than the 1 mm depth taken from cobbles for conventional processing (Simms et al. 2011).

Another consideration is that measurements made at the 1 mm depth still result in power densities (from 95 W/cm^2 for the IR laser to 355 W/cm^2 for the green laser) that are more than double the conventional instrument's 50 W/cm^2 incident power. It is possible that the lasers will continue to stimulate luminescence from the sample at the calculated penetration depths and deeper, and further measurements are required to truly understand the effects of laser stimulation at sample depths below 1 mm. Recommendations for the execution of these measurements are discussed in chapter 9.

Chapter 7

X-Ray Characterization

Chapter 3 and chapter 4 described the necessity of a calibration radiation source for developing spatially-resolved dose maps of sample surfaces. For this purpose, a Magnum 10-50 kV, 5-200 μ A, X-ray source with a silver target and equipped with a 50 μ m aluminum filter and 18 mm brass collimator was selected. While the laser stimulates the sample point by point, it is desirable to irradiate the entire sample surface at once to reduce the measurement time. In order to provide accurate irradiations, the dose rate of the source must therefore be uniform, or any non-uniformities must be well characterized. Non-uniformities could arise from dose rate dependencies on sample composition, lateral position from the center of the beam, inherent X-ray tube stability, X-ray energy, and the depth of dose deposition. This chapter presents the methods and results of characterizing the X-ray source for use with spatially-resolved dose mapping. The first section presents the methods that were used to characterize energy output and uniformity of the X-ray source, and the calibration of the X-ray source against the Risø beta source is described in the second part of this chapter.

7.1 Energy Spectrum and Source Filtration

It has been found that the luminescence response of some materials is energy dependent for low X-ray energies (Hong et al. 2005; Thomsen et al. 2006). In addition, different X-ray energies have different penetration depths. This causes the applied dose to be depth-dependent and causes large uncertainties with source calibration. It is therefore recommended to filter the X-ray source to harden the spectrum by removing low-energy radiation.

An AmpTek XR-100-CdTe X-ray detector was used to measure the energy spectrum of the tube with varying filters. Six filters of varying thickness and material were acquired from AmpTek as a kit for use with LuCIDD's X-ray source for energy spectrum analysis: 250 μm Al, 1 mm Al, 25 μm Ag, 25 μm Cu, 25 μm Mo, and 25 μm W (Fig. 7.1). A custom, 50 μm Al filter was purchased separately. The 6.35 mm diameter circular filters were adhered directly to the X-ray source's nozzle using tape for energy spectrum measurement. All measurements were conducted at a source-to-detector distance of 28.6 cm for an irradiation time of 60 seconds and at 50 kV and a reduced current of 40 μA to prevent saturation of the detector. Figure 7.2 shows an image of the experimental setup. Prior to measurements, the detector had to be calibrated. The procedure for calibrating the detector and measuring the spectra can be found in appendix E.

A background measurement was made with the tube switched off. Figure 7.3a shows the spectrum of the unfiltered X-ray tube and the spectrum of the source after passing through the 50 μm aluminum filter that was chosen for this work. It can be seen in figure 7.3 that the energy goes to zero at values below a certain threshold. This is because the noise at those energies was enough to cause the detector to "pile-up," i.e., it became saturated, which prevented accurate measurement of the spectra. To combat this, a large slow-threshold was required at 24%. Because of the limitations of the detection instrument, the 50 μm aluminum filter was selected primarily due to the recommendation



Figure 7.1: The six filter types acquired in AmpTek's Mini X-ray Filter kit. Each filter was 6.35 mm in diameter.

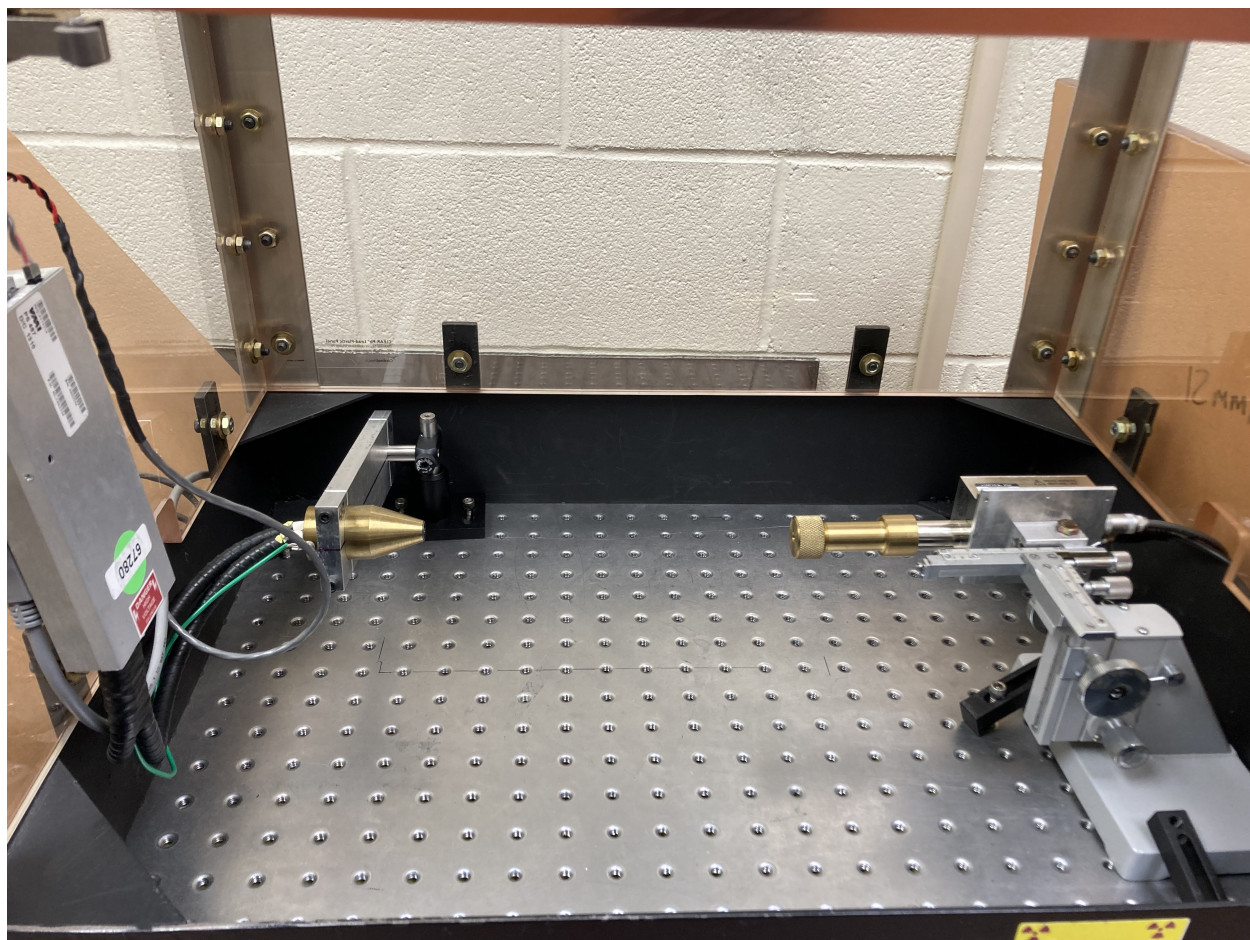


Figure 7.2: Varying filters were applied to the X-ray source (left), and it was directed at the X-ray detector (right), which is equipped with a 4.45 cm brass collimator.

made by Hong et. al. (2005).

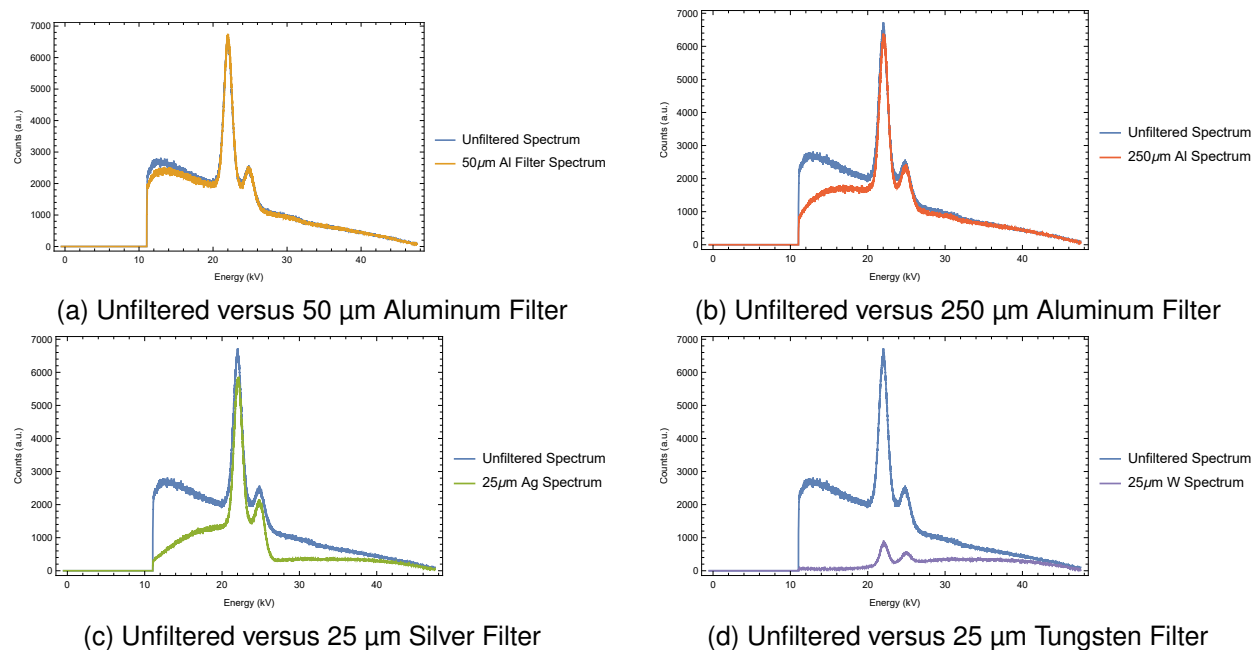


Figure 7.3: Comparisons between the unfiltered and filtered spectrum of the X-ray tube for four filters. The filter in (a) was selected for the work presented here.

7.2 Beam Uniformity

GAFChromic EBT-3 radiochromic film was used to measure the spatial uniformity of the X-ray tube's output beam. EBT-3 is made up of an active film layer that is inserted between two layers of clear, 125 μm polyester. When the film is irradiated, it begins to darken at a rate that is proportional to the incoming radiation. Irradiated films are scanned using a high-resolution scanner and imported into ImageJ to measure the darkness value of each pixel. The darkness values were calibrated to applied dose using a known X-ray source. The values give a direct measurement of the dose applied by LuCIDD's X-ray tube. The position and applied dose of each pixel is imported into Mathematica for analysis. Details of the GAFChromic calibration, and making measurements with the film can be found in appendix E.

A set of films were cut into fifteen 2 x 2 inch squares, placed into the sample tray, and irradiated using LuCIDD's X-ray source for varying times. All irradiations were performed at 50 kV and 200 μ A with a 50 μ m aluminum filter and an 18 mm brass collimator. Figure 7.4 shows a 3-D representation of the normalized dose relative to the position of each pixel for a scanned film.

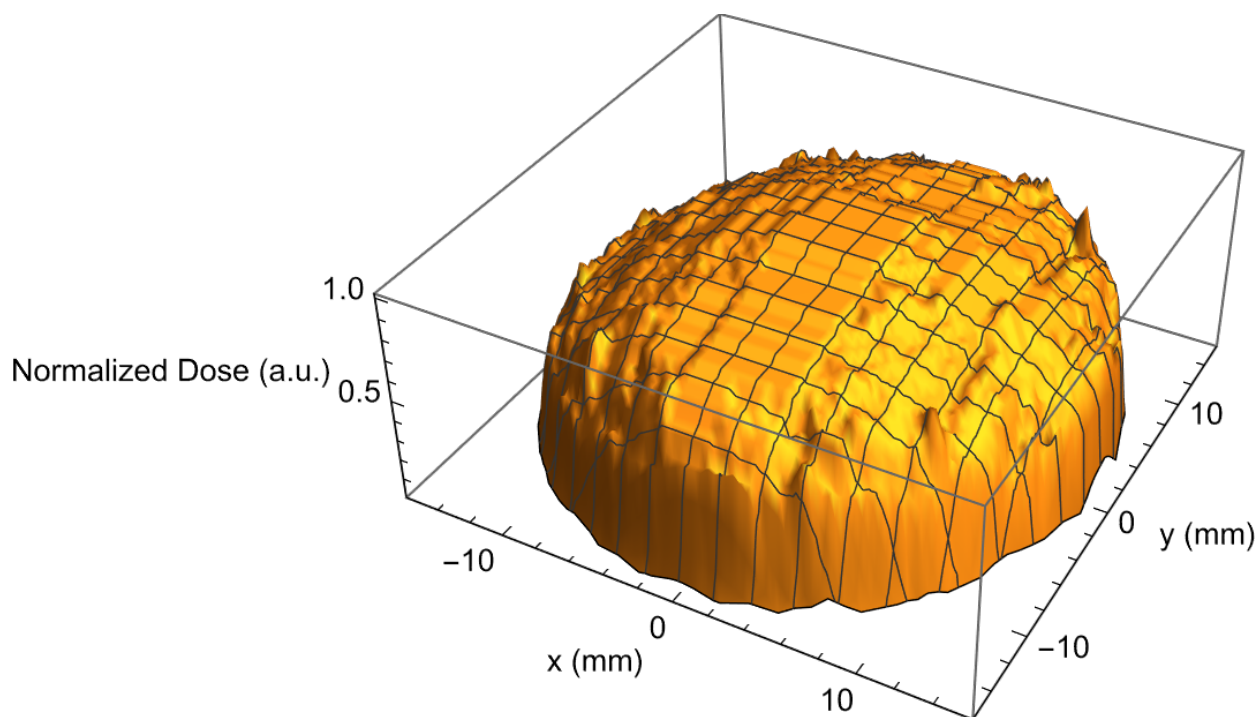


Figure 7.4: Using ImageJ, the darkness values of the pixels are converted to dose and imported into Mathematica for visualization and analysis.

A circular area around the center of the beam was taken at different radii for each film. The dose rate of each pixel within the area was averaged and normalized to the mean dose rate from the 6.45 mm diameter measurement (2.03 cGy/s). The circle's diameters were chosen to coincide with the sizes of standard samples, (e.g., Luxel dosimeters or Risø sample cups), as well as the full 18 mm collimator of the X-ray tube. Table 7.1 shows the normalized dose rates and relative standard deviations for the chosen sample diameters. For the 18 mm sample size, the mean dose rate decreased by 6.4%; however, the relative standard deviation from the mean is only 1.30%. For the 9.67 mm rock

slices used thus far, the mean dose rate only decreased by 2% of the maximum, and the variation was only 0.63%.

Table 7.1: Relative standard deviations from the average dose for four diameter break-points.

Selected Sample	Beam Diameter (mm)	Normalized Dose Rate (a.u.)	Relative Standard Deviation (%)
Luxel	6.45	1	0.53
Risø Cup (Inner Diameter)	8.40	0.995	0.60
Rock Slice	9.67	0.990	0.63
Risø Disk	11.65	0.980	0.73
X-Ray Collimator	18.00	0.936	1.30

7.3 Dose Linearity

Samples used for dating have a large variety of dose values. For younger samples with low doses, uncertainties are introduced by the short irradiation times. Because of this, it is desirable to have a source with a low dose rate so that the irradiation time can be increased. X-ray sources have a variable dose rate that is controlled by adjusting the tube current. The connection between current and dose rate is expected to be linear (Andersen, Bøtter-Jensen, and Murray 2003; Thomsen et al. 2006).

The linear response of the dose rate relative to the current of the X-ray tube was characterized using GAFChromic film. A series of ten measurements were taken at 50 kV for 180 s and with tube currents varying from 20 μ A to 200 μ A. The dose rates were normalized relative to the maximum dose rate at 200 μ A (1.77 cGy/s), and a linear fit was applied to the data (Fig. 7.5). The results show an X-ray source with remarkable linearity as the relative standard error of the slope of the fit line is 1.31%.

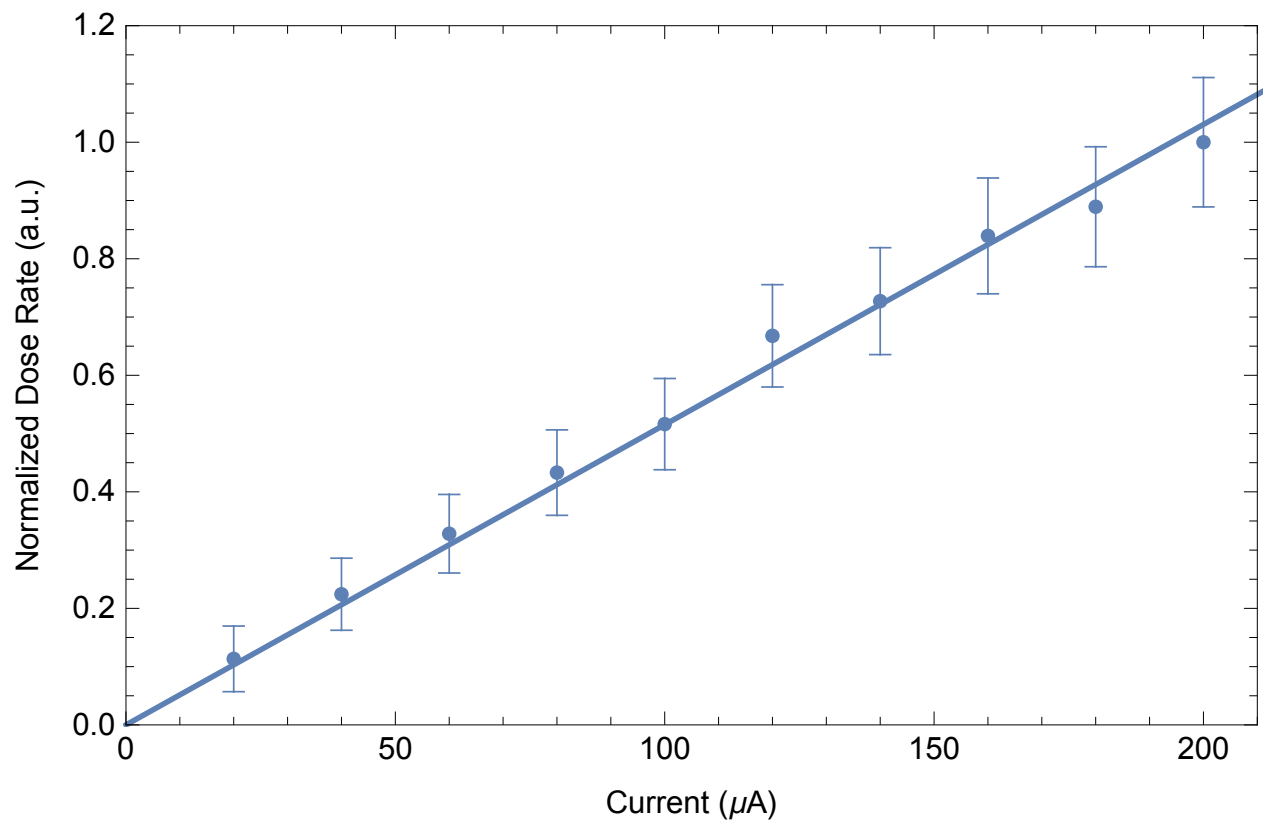


Figure 7.5: The count rate of the X-ray source increases linearly with tube current.

7.4 Dose Rate

In order to convert data from a luminescence signal to an equivalent dose, the dose rate of the luminescence reader's radiation source must be known. Gamma and beta sources are considered to be the standard in luminescence dosimetry because luminescence efficiency is constant over the range of natural beta and gamma energies. As mentioned earlier in this chapter, the luminescence efficiency of some materials varies for the lower range of X-ray energies. Because of this, LuCIDD's X-ray source was calibrated against the beta source of the Risø TL/OSL DA-20 luminescence reader. The Risø reader is usually used for mineral separates and not for the mineral mixtures encountered in rocks, so the beta source was first calibrated for these materials. This calibration was performed by applying the SAR procedure which has been outlined in chapter 3. The following section describes the calibrations of the Risø's beta source and LuCIDD's X-ray source.

7.4.1 Calibration of the Risø Reader's Beta Source

The four materials used in this research are quartz sediment, quartzite rock slices, sandstone rock slices, and Luxel[®]+ dosimeter tape ($\text{Al}_2\text{O}_3\text{:C}$). Luxel dosimeter tape was obtained from Landauer as a reel (1 m long, 20 mm wide, 0.3 mm thick). It consists of $\text{Al}_2\text{O}_3\text{:C}$ grains sandwiched in a thin film between two layers of laminate. The aluminum oxide crystals are grown by Landauer and ground into powder (20 - 90 μm) to produce these dosimeter tapes (Yukihara and McKeever 2008). As mentioned previously, the Risø's beta source had already been calibrated for Luxel dosimeter tape, as well as quartz grains.

In the research presented here, the Risø's beta source was calibrated for the quartzite and sandstone rock slices and also, for comparison, 90 μm - 125 μm quartz grains. The sediment, rock slices, and dosimeter tape were each prepared using separate methods.

They were tested for suitability for luminescence measurements and their ideal measurement parameters were determined. A last test ensured that a known applied dose can be measured accurately. Once prepared, the samples (except Luxel) were packaged in vials sent for calibration irradiations with a known ^{60}Co source. Details of these steps can be found in appendix E.

Due to the limited number of aliquots produced for each sample, only a few different doses were applied: quartz sediment received doses of 5 and 7 Gy, the quartzite rock slices received doses of 5, 7, and 10 Gy, and the sandstone rock slices received doses of 5 and 7 Gy.

Each calibration sample was divided into aliquots. The SAR procedure was used to determine the beta irradiation time that corresponds to the dose absorbed by the respective aliquot. The applied calibration dose in Gy was then divided by the measured beta irradiation time to determine the dose rate of the beta source. The results for 31 aliquots of quartz sediment are shown in figure 7.6. The beta source calibration dates and dose rates for each sample may be found in table 7.2, along with those for the X-ray source described in the next section. The value for Luxel was not measured here but was added to the table for the sake of completeness.

Table 7.2: Risø beta source and X-ray source dose rates in different samples. Note, the X-ray source was calibrated relative to the Risø beta source prior to the Risø's calibration to the ^{60}Co source.

Source	Sample	Dose Rate (mGy/s)	Calibration Measurement Date
$^{90}\text{Sr}/^{90}\text{Y}$ Beta	Quartz Sediment	81.7 ± 0.4	March 7, 2023
$^{90}\text{Sr}/^{90}\text{Y}$ Beta	Quartzite Rock Slices	78.68 ± 0.28	March 14, 2023
$^{90}\text{Sr}/^{90}\text{Y}$ Beta	Sandstone Rock Slices	83.2 ± 0.6	March 9, 2023
$^{90}\text{Sr}/^{90}\text{Y}$ Beta	Luxel	72.7 ± 1.5	April 23, 2021
50 kV X-ray	Quartz Sediment	79.7 ± 1.0	November 8, 2022
50 kV X-ray	Quartzite Rock Slices	71.5 ± 0.8	November 8, 2022
50 kV X-ray	Sandstone Rock Slices	77.8 ± 0.5	November 30, 2022
50 kV X-ray	Luxel	123.0 ± 0.8	November 15, 2022

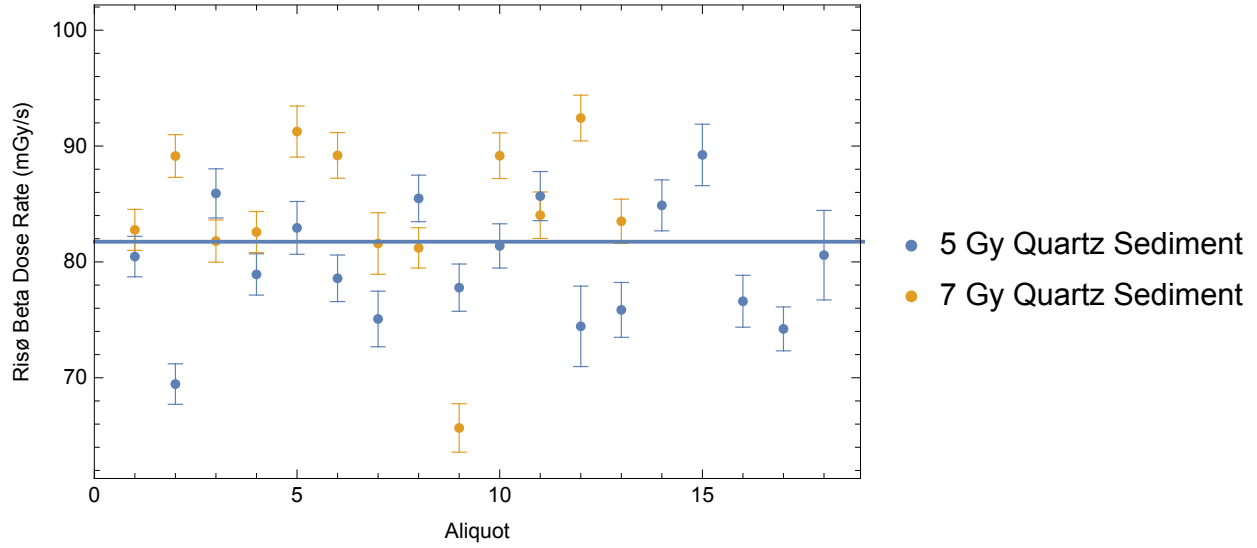


Figure 7.6: The Risø beta source was calibrated for dose rate in multiple sample types. This plot shows the beta dose rate calculated from measuring aliquots from two sets of quartz sediment.

7.4.2 Calibration of the X-Ray Source

Whether it's X-rays or beta irradiation, it is important to calculate the given dose relative to a calibrated, standardized source. The majority of standardized sources are calibrated against an ionization chamber which provides the energy transferred to the target per unit mass (kerma) and is measured in Gy (Attix 2004). The dose applied to a sample, in this case quartz, can be determined using:

$$D_Q = K_{air} \left(\frac{\mu_{en}}{\rho} \right)_Q / \left(\frac{\mu_{en}}{\rho} \right)_{air} \cdot A \quad (7.1)$$

D_Q is the dose to quartz, K_{air} is the air kerma as measured by the ionization chamber, (μ_{en}/ρ) are the mass-energy absorption coefficients for quartz and air and A is the attenuation through the sample and its storage vial:

$$A = e^{-(\mu/\rho)_Q \rho_Q L_Q} e^{-(\mu/\rho)_v \rho_v L_v} \quad (7.2)$$

with (μ/ρ) as the mass attenuation coefficient. ρ as the density of the material and L as

the effective attenuation path length. The mass-energy absorption coefficient for quartz (SiO_2) in equation 7.1 is calculated simply by summing the coefficients of the constituent atoms relative to their mass ratios:

$$\left(\frac{\mu_{en}}{\rho}\right)_Q = \frac{m_{Si}}{m_Q} \left(\frac{\mu_{en}}{\rho}\right)_{Si} + \frac{2m_O}{m_Q} \left(\frac{\mu_{en}}{\rho}\right)_O \quad (7.3)$$

The dose rate of LuCIDD's X-ray source was calibrated for four samples: quartz sediment, quartzite rock slices, sandstone rock slices, and Luxel dosimeter tape. Multiple aliquots of each sample were irradiated by the X-ray source for different times, ranging from 10 to 300 seconds. All measurements were performed at 50 kV and 200 μA with a 50 μm aluminum filter and 18 mm brass collimator. The SAR procedure was performed on the irradiated aliquots using the Risø reader to determine their resulting equivalent dose of beta in Gy. A linear fit was applied to this data to determine the dose rate of the X-ray source in each material (Fig. 7.7). Table 7.2 shows the dose rates of the X-ray source for each sample material.

Considerations for Luxel Al_2O_3 Dosimeter Tape

$\text{Al}_2\text{O}_3\text{:C}$ bleaches very slowly and the Luxel cannot be heated so they must be re-set optically. Therefore, the current methods for measuring Luxel dosimeter tape with the Risø require ten minutes of stimulation, six times longer than measurements for other materials. Usually, a dose response curve is built for every single aliquot individually, since natural materials are non-uniform and show large inter-aliquot variation. Luxel, however, is artificially produced, and consists of uniform material. To circumvent compounding measurement times, a single calibration curve was built for the Luxel dosimeter tape (Yukihara et al. 2008). Twenty-one aliquots were given known doses from 0.727 to 5.45 Gy and were measured for 600 seconds. Each aliquot was then given a test dose of .727 Gy and measured for another 600 seconds. The resulting calculated signal ratio (L_x/T_x) pro-

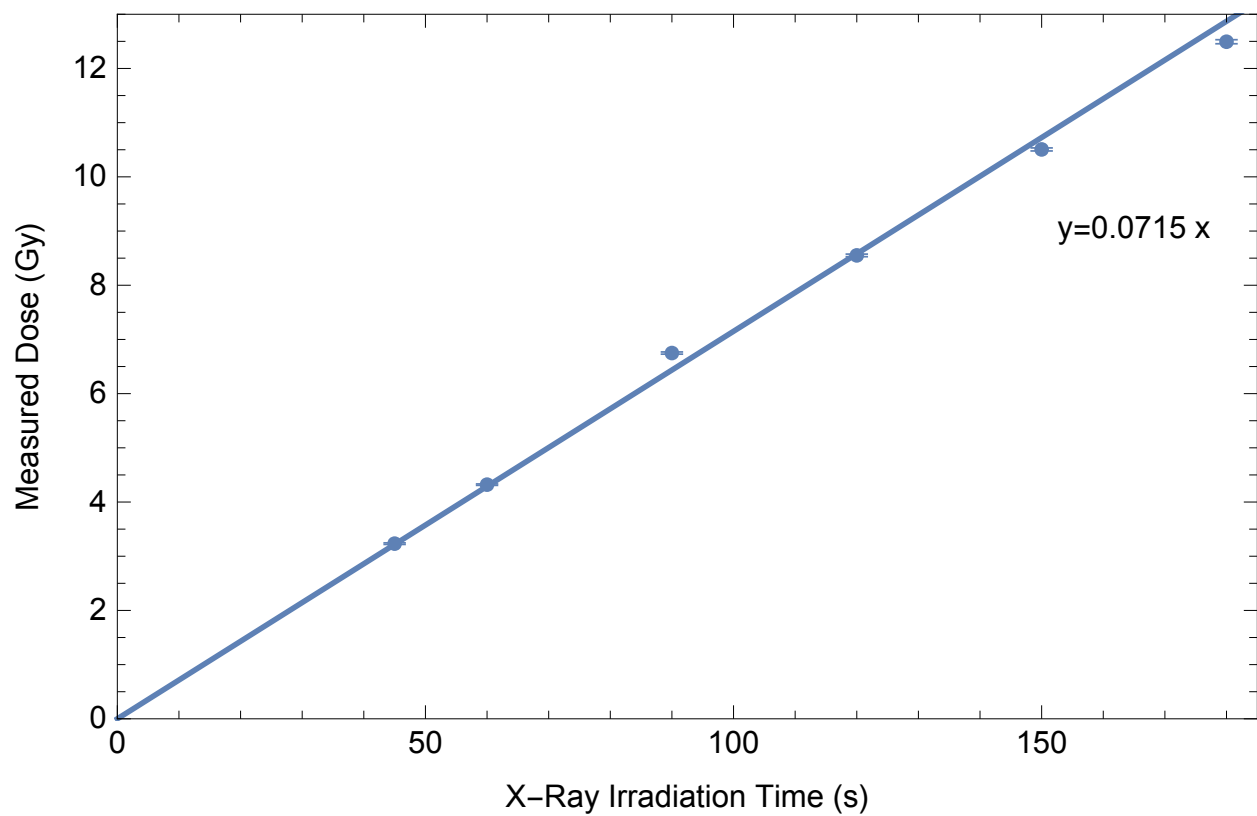


Figure 7.7: Plot of the X-ray dose measured by the Risø in quartzite rock slices versus the X-ray irradiation time. The slope of the fit line is the dose rate of the X-ray source in quartzite slices.

vided a linear response to the applied dose (Fig. 7.8). All future measurements for Luxel were performed in similar fashion, with each aliquot having its “natural” and test signals measured a single time - without the need for multiple regeneration points as is usually required - and the signal ratios were placed on the fit curve to determine the applied dose. This reduced measurement times from about two and a half hours per aliquot to about twenty minutes per aliquot.

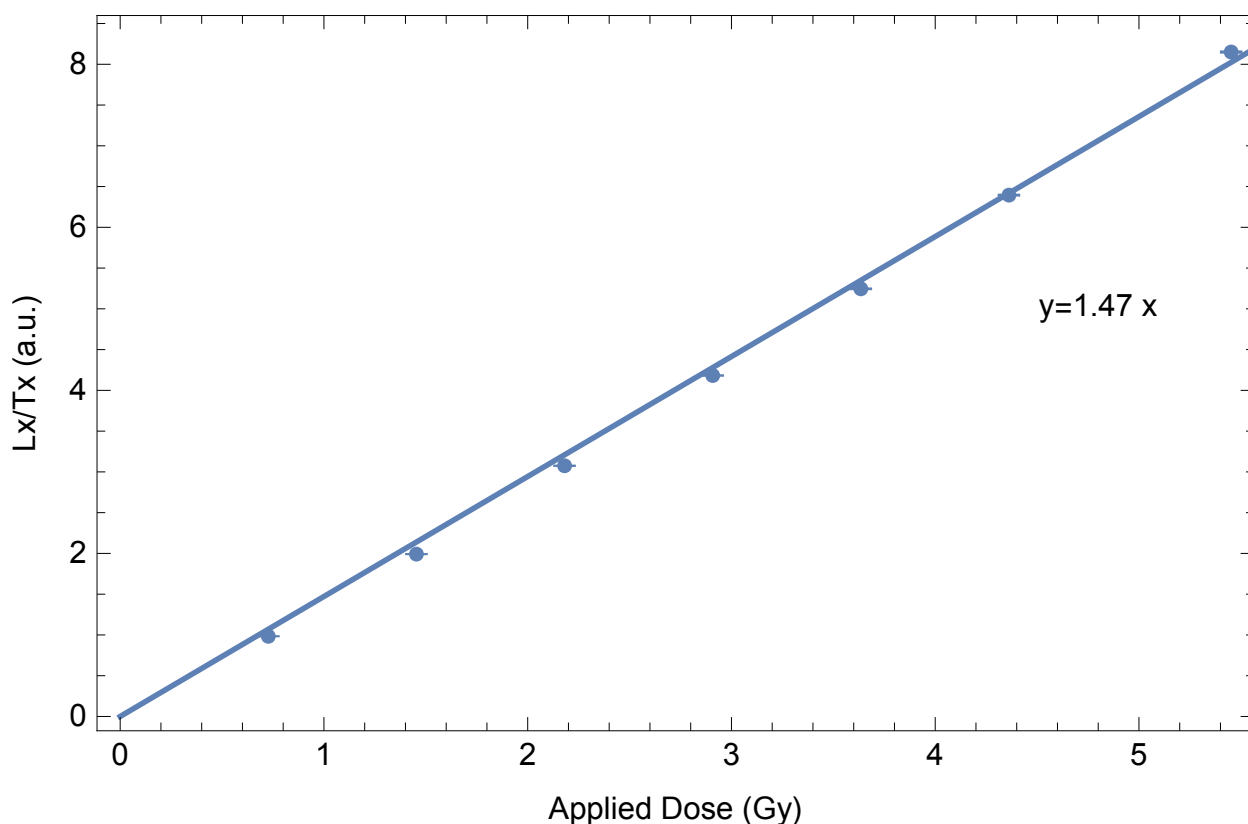


Figure 7.8: Luxel’s signal response to the Risø’s beta radiation source.

7.4.3 Warm-up Time and Stability

One possible source of uncertainty with an X-ray source is the temperature dependence of the output. X-ray sources require warm-up time, and the current is temperature dependent. Therefore, the short-term stability of LuCIDD’s X-ray source was determined during and after warm-up. Twenty-three aliquots of quartz sediment were irradiated with

the X-ray source back-to-back for 180 seconds each. All irradiations were applied at 50 kV, 200 μ A and with a 50 μ m aluminum filter and 18 mm brass collimator. The aliquots were measured in the Risø TL/OSL reader, and the SAR procedure was used to determine the dose applied to each aliquot. The measured doses were normalized to the average dose of the dataset (Fig. 7.9). The relative standard deviation from the mean dose is 4.01% with little difference between initial (cold) and final (warm) irradiations.

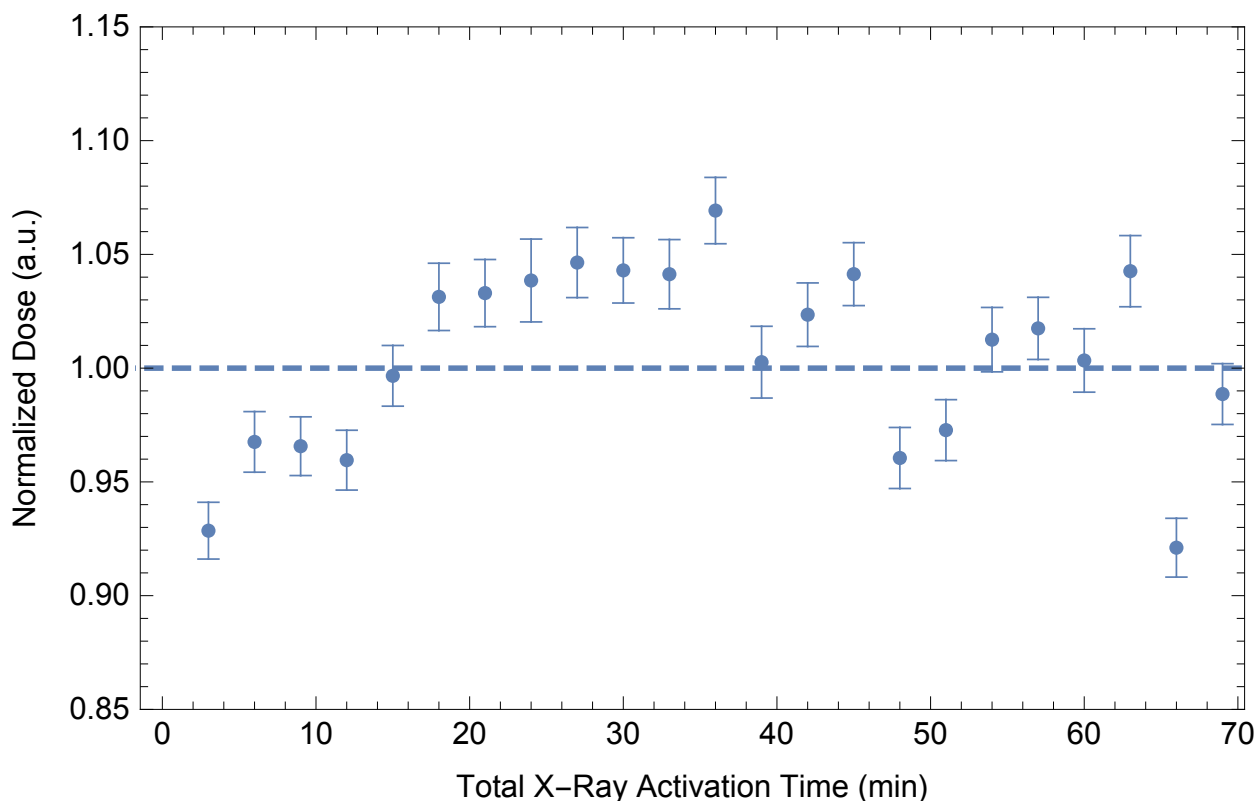


Figure 7.9: Applied dose relative to total X-ray activation time. The applied dose rate has a relative standard deviation from the mean of 4.01%, with all values within 10% of the average.

7.4.4 Penetration Depth

To measure the penetration depth of the X-ray source, aliquots of quartz sediment and Luxel dosimeter tape were placed beneath slices of granite with thicknesses varying from 0 to 3.85 mm. The aliquots were irradiated, and their applied dose rates were measured

using a Risø TL/OSL reader. The resulting doses were normalized to the zero-depth dose rate. A curve was fit following the form of the Lambert-Beer law (Fig. 7.10). The penetration depth was determined as the sample depth where the applied dose rate falls to $1/e$ of the normalized dose. Using quartz sediment and Luxel dosimeters, the penetration depth of X-rays in granite was found to be 0.46 ± 0.04 mm and 0.42 ± 0.06 mm, respectively, providing an average penetration depth of 0.447 ± 0.035 mm in granite.

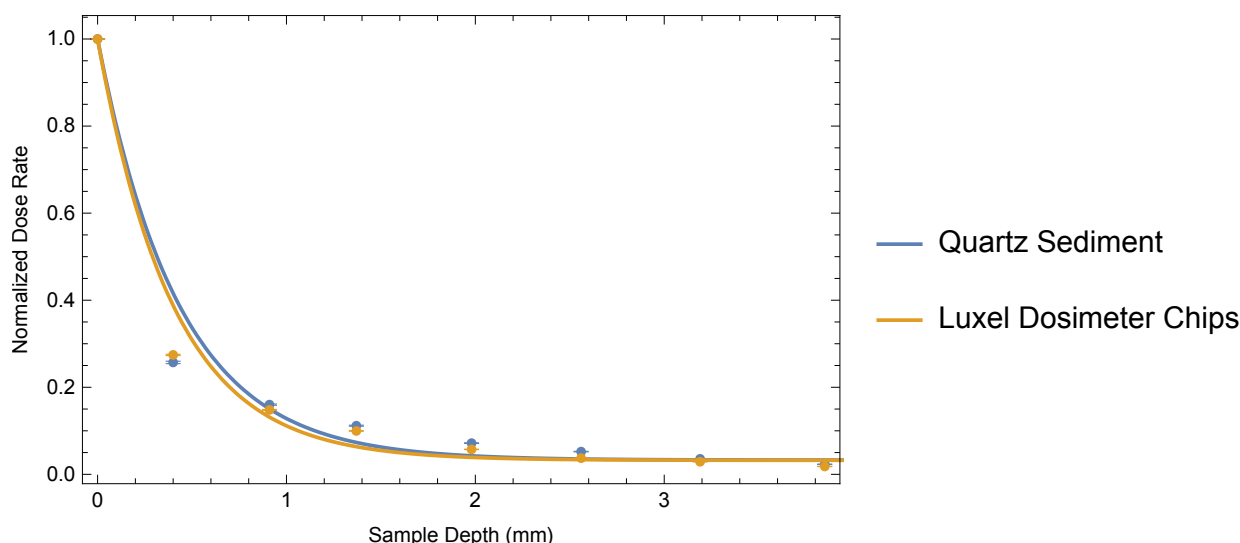


Figure 7.10: Combined Penetration Depth

7.5 Discussion

The energy spectrum of the X-ray tube was measured with and without multiple filters. Little change in the spectrum was observed between the unfiltered and the 50 μ m aluminum filtered spectra. Hong et. al. (2005) saw a similar slight change in spectra when compared with thicker absorbers, and the largest drop in X-ray intensity was found at energies lower than 10 kV. Because of this, it is possible that there is a larger effect of the aluminum filter on the lower energies which were cut off during our measurements. Hong et. al. also found a large decrease in the standard error of the resulting signal when irradiating with a 50 μ m aluminum filter when compared to dose applied with no

filter. Thus, a filter of the size used in this work (50 μm) has been deemed sufficient to decrease uncertainty in the signal collection resulting from irradiation without dramatically reducing the dose rate of the source. Dose rates of the X-ray source were determined only for the 50 μm aluminum filter, but other filters might produce comparable dose rates while hardening the spectrum more than the 50 μm aluminum filter.

The spatial variation of the X-ray source's dose rate across a sample surface was measured and the relative standard deviation from the mean applied dose was calculated at four diameters. The relative standard deviations were all less than 5% within a sample diameter of 18 mm – the size of the X-ray source's collimator. This spatial uniformity is exceptional when compared to the conventional beta source's. Spooner and Allsop (2000) found that their beta source had a relative standard deviation of around 15% across a 10 mm sample area with a source-to-sample distance of 15 mm. Note that this source-to-sample distance is over twice the distance that is normally used for conventional Risø readers and thus, the conventional uniformity is thought to be worse across the standard 10 mm sample size.

The dose rate of the X-ray source displayed excellent linearity relative to the applied tube current. This will allow irradiations at lower dose rates which is useful for reducing error in the equivalent doses of young, low dose samples.

The observed dose rates for the X-ray source in Luxel dosimeters, quartz sediment, quartzite rock slices, sandstone rock slices were found to be 123.0 ± 0.8 mGy/s, 79.7 ± 1.0 mGy/s, 71.5 ± 0.8 mGy/s, and 77.8 ± 0.5 mGy/s, respectively. The X-ray dose rates in sediment and rock slices are comparable to those applied by the Risø, as they are all within 10% of its beta source's dose rate. The X-ray source's dose rate was much higher than the beta's in Luxel, indicating an increased response of $\text{Al}_2\text{O}_3:\text{C}$ to X-ray energies.

It must be noted that while the applied doses of the Risø and X-ray are comparable, the dose rate calibration of the Risø's beta source resulted in a sediment dose rate that was about 20% lower than what was expected. At this point, the reason for this is not fully clear.

It is possible that the dose quoted by the gamma irradiation facility was incorrect. Any error in the Risø source calibration will carry over to the X-ray source. These calibration measurements for the Risø are planned to be repeated in the future using a different source.

No dose rate dependence on operation time was observed for the X-ray source, with a relative standard deviation of only 4.01% across 70 minutes of irradiations. This allows us to disregard warm-up time with respect to the X-ray source and allows for faster measurement sequences overall.

The penetration of the X-ray source into granite was measured to be 0.447 ± 0.035 mm. This value is between the top and bottom end of the laser penetration depths. This introduces another possible source of signal confusion, as lasers centered on translucent areas of a sample will penetrate beyond the X-ray penetration depth, stimulating the full extent of deposited dose. Lasers targeting opaque grains won't be able to reach these depths to stimulate the full deposited dose, and thus, will have less signal to show for it. This disparity could cause fluctuations in the dose map that are not caused naturally and should be further investigated. A discussion on proposed future research to refine characterizations of the X-ray source may be found in chapter 9.

Chapter 8

Testing LuCIDD for Spatially Resolved SAR

In chapter 4, we discussed the requirements to perform spatially resolved dosimetry and dating measurements on whole rock surfaces using optically stimulated luminescence. Chapter 5 described an instrument complete with all required components for performing the SAR procedure, and chapter 6 and chapter 7 characterized these components so they may be more accurately applied to develop dose maps of sample surfaces.

Before full dose maps can be created, LuCIDD - as a complete instrument - must be characterized and tested. It is necessary to prove that LuCIDD's current state is fully capable of providing accurate dosimetry measurements. Spatially resolved dose maps have yet to be produced, but this chapter presents the characterizations that have been performed to enable these future measurements.

This chapter describes the focusing process for the microscope objective. The instrument is characterized for the minimum acceptable stimulation time, and LuCIDD's ability to perform the SAR dosimetry procedure is determined. The spatial resolution of the dose map measurement is equivalent to the size of the individual, resolvable pixels. These pixel sizes are affected by laser focal spot size, laser power density, and minimum re-

quired stimulation time. Attempts to determine the resolution of LuCIDD are summarized in the last part of this chapter.

8.1 Methods

8.1.1 Details of the SAR Procedures

Dose recovery tests were performed on Luxel dosimeter tape and quartzite rock slices to determine LuCIDD's ability to perform the SAR procedure and recover a known applied dose. Luxel dosimeter tape was also used to determine the minimum stimulation time and the resulting resolvable pixel size. The preparation of these samples follows the same process as was described in chapter 7 and appendix E.3.

LuCIDD's green laser, green dichroic mirror, and a 330 WB80 detection filter were used to stimulate and collect luminescence from Luxel dosimeter tape (Yukihara et al. 2008). For quartzite rock slices, the green laser, blue dichroic mirror, and a UG 11 detection filter were used. The green laser was chosen for quartz instead of the blue laser in an attempt to duplicate stimulation methodologies from previous research (Duller et al. 1999; Ballarini et al. 2005; Greilich, Glasmacher, and Wagner 2005; Jacobs et al. 2013). Fig. 8.1 shows the laser emission wavelength, and the transmission wavelengths of the chosen filters. Thus far, LuCIDD has utilized only the green laser to stimulate Luxel dosimeter tape (330 WB80 filter) and quartzite rock slices (UG11 filter). While it appears that both filters transmit luminescence in similar bands, the wider transmission band and the greater overall transmittance of the UG11 filter allows the dimmer quartzite samples to be measured more clearly. Owing to its narrower transmission band, the 330 WB80 filter is used to remove the time-dependent UV component of Luxel luminescence that resides at wavelengths <330 nm (Yukihara et al. 2008).

All irradiations were performed at 50 kV, 200 μ A, with a 50 μ m aluminum filter and

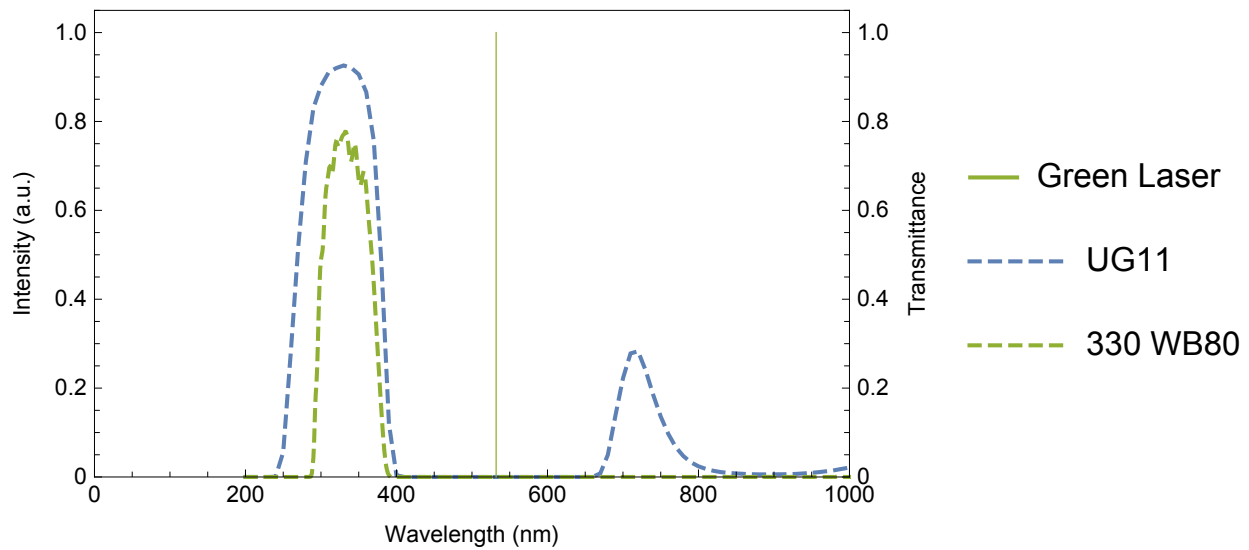


Figure 8.1: Emission wavelength of LuCIDD's green laser and transmission wavelengths of LuCIDD's detection filters.

an 18 mm brass collimator. The quartzite aliquots received preheat treatments during measurement of 200°C which were held for 60 seconds, cutheats of 160°C, and were measured at 125°C. The sample stage was cooled to 60°C after each heating. No heat treatment was performed on the Luxel aliquots to prevent damaging the temperature-sensitive polyester sheets of the dosimeters. Tables 8.1 and 8.2 show the SAR procedure for Luxel and quartzite rock slices, respectively.

Table 8.1: Steps of the SAR cycle for Luxel dosimeter tape used with LuCIDD.

Step	Action
1	Irradiate with a dose
2	Measure OSL for 10s
3	Irradiate with test dose
4	Measure OSL for 10s
5	Repeat steps 1-4 for each regeneration and test point

LuCIDD's objective was focused onto aliquots using the non-stimulating red laser and beam splitter. The SAR procedure was performed on the aliquot using the green laser for

Table 8.2: Steps of the SAR cycle for quartzite rock slices used with LuCIDD.

Step	Action
1	Irradiate with a dose
2	Preheat for 60 s at 200°C
3	Cool to 125° and Measure OSL for 10s
4	Irradiate with test dose
5	Cutheat at 160°
6	Cool to 125° and Measure OSL for 10s
7	Repeat steps 1-6 for each regeneration and test point

stimulation, the X-ray source for calibration irradiations, the heating plate for temperature control, and the photomultiplier tube (PMT) for signal collection. The signal data was analyzed using the **R** “Luminescence” package to provide equivalent doses which were imported into Mathematica for analysis and visualization.

8.1.2 Focusing Method

As discussed in chapter 5, confocal instruments measure only light that is emitted in the focal point of the microscope objective. Light from outside the focal point is blocked by the detection pinhole. Rock surfaces are rough and uneven; the optimum distance between objective and sample is therefore different for each stimulated spot on the sample and must be determined prior to measurement. Focusing is usually performed by reflecting a laser off the surface of a sample and maximizing the reflected intensity. However, with OSL measurements, any light-exposure has the potential to de-trap electrons and to stimulate luminescence. Red wavelengths were found to have very little to no effect on OSL samples (Aitken 1998).

All four lasers were focused onto the same spot of the same sample using the focusing procedure described in section 5.2.1. The orange curve in figure 8.2 shows the intensity of the reflected red laser versus the objective position.

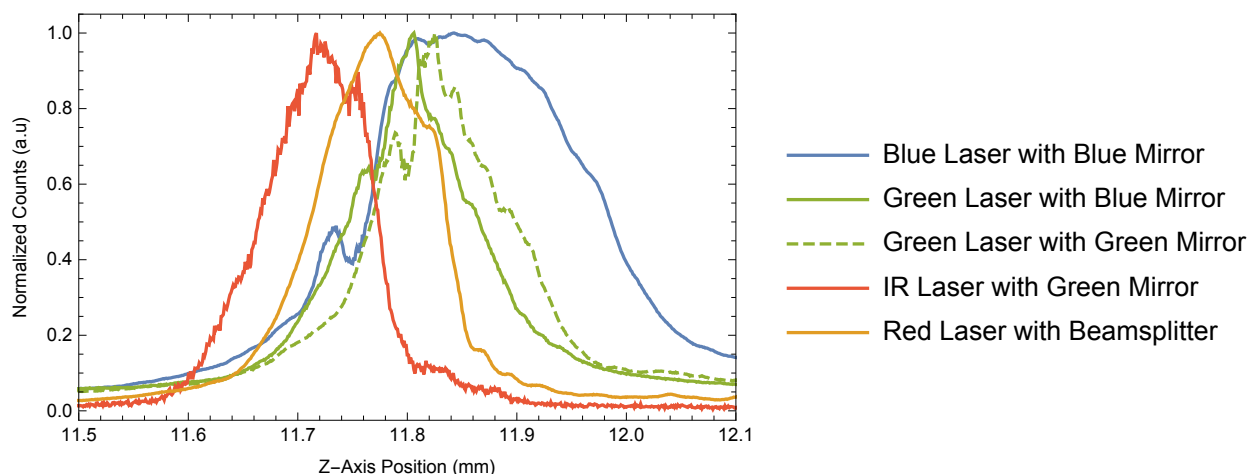


Figure 8.2: Reflected laser intensity versus microscope objective position. Using the same sample, each laser and mirror combination was scanned along the entire z-axis (0 to 25 mm). Each combination of laser and dichroic mirror produced curves of different shapes and widths.

A gaussian curve was fit to the recorded PMT data using Mathematica and the z-axis position that correlated to the maximum laser transmission was found. This position represented the focal length of the microscope objective to the sample and corresponded to the minimum spot size of the laser on the sample. Since it is crucial that the focal spot of the red laser coincides with the focal spot of the stimulation laser, other lasers were tested as well. Figure 8.2 shows reflected intensity for multiple laser and mirror combinations striking the same sample. Different combinations provided different curve shapes and widths. Due to small differences in the alignments of the lasers and confocal dichroic mirror assembly, there are small differences in z-axis position. Table 8.3 shows these offset values. The measurement procedure for a dating sample is therefore adjusted as follows: For every point on the surface of a sample, the focal length (i.e., the z-axis position) is first found using the red laser and beamsplitter. This process of scanning and analysis is repeated for every desired (x, y) pixel on a sample to provide their corresponding z-axis focal lengths. For subsequent stimulation with another laser and mirror combination, the known offset value is added to the measured z-axis position.

Table 8.3: Focal length offsets relative to the red laser and beamsplitter.

Laser	Dichroic Mirror	Focal Offset (mm)
Red	Beamsplitter	-
Blue	Blue	+0.091
Green	Blue	+0.033
Green	Green	+0.059
IR	Green	-0.053

8.2 Stimulation Time

One important factor in the SAR procedure is the stimulation time, i.e., the time for which the light source is switched on and the luminescence signal is recorded. In the SAR procedure the same sample is measured multiple times and it is crucial that the sample be completely reset during stimulation. On the other hand, long stimulation times lead to a long - and possibly impractical – overall duration of a measurement. Stimulation times should therefore be optimized.

LuCIDD was used to determine the shortest effective stimulation time to acquire signals that produce accurate equivalent doses (within 10% relative standard error). Three sets of three Luxel aliquots were irradiated with 5.04 Gy of X-ray and measured by LuCIDD using varying stimulation times of 2, 5, and 10 seconds. Again, only single points were measured on each aliquot. The ratio of the recovered dose to the applied dose was then plotted against the stimulation time (Fig. 8.3). The stimulation times, dose ratios, and relative standard errors are reported in table 8.4. A time of 10 seconds was selected as the minimum required stimulation time, though different times may be suitable for different materials.

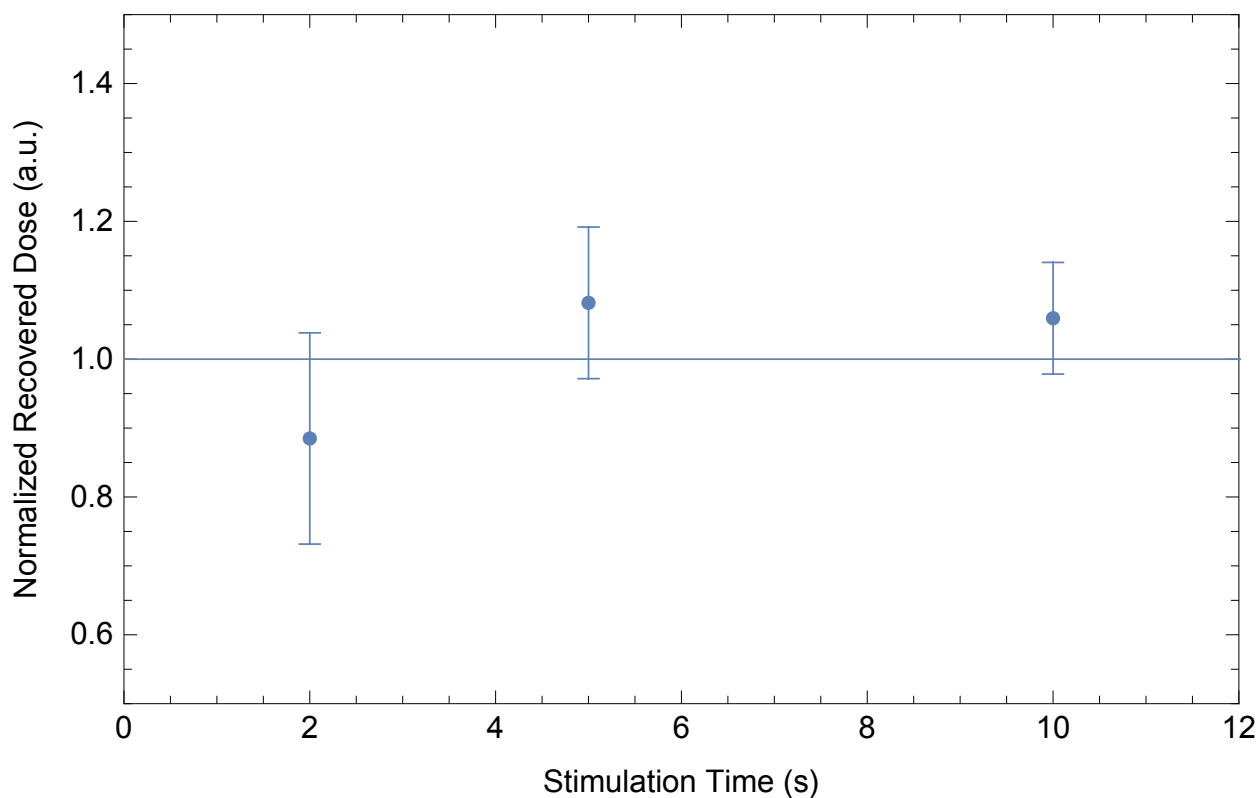


Figure 8.3: The relative standard deviation decreases with increasing stimulation time. The horizontal line represents a perfectly recovered applied dose. The error bars represent the standard error for three aliquots.

Table 8.4: Ratio of recovered dose and applied dose for varying stimulation times. Relative standard errors were calculated from three aliquots for each applied dose.

Stimulation Time (s)	Dose Ratio	Relative Standard Error (%)
2	0.88 ± 0.15	17.32
5	1.08 ± 0.11	10.17
10	1.06 ± 0.08	7.65

8.3 Dose Recovery Tests

The overall purpose of LuCIDD is to accurately measure the radiation dose absorbed by minerals. The simplest and most commonly used strategy to test the performance of a luminescence instrument is to irradiate a sample externally with a known dose and to measure this dose with the instrument. Two dose recovery tests were performed to evaluate LuCIDD's ability to recover a known dose, and thus its ability to perform the SAR procedure on a sample with an unknown natural dose. These tests were performed using Luxel dosimeter tape and quartzite rock slices. No dose maps were made during these tests, as only a single point was measured on each aliquot.

For conventional SAR procedures performed using the Risø luminescence reader, a difference of 10% between the applied and recovered doses is deemed acceptable for natural samples (Wintle and Murray 2006). Due to LuCIDD's confocal detection pinhole, it is expected that the reduction in signal could be crippling for the instrument's ability to provide stored dose information.

Various known doses of up to 5.04 Gy were applied to six Luxel aliquots and doses of up to 12.87 Gy were applied to four quartzite aliquots. The SAR procedure was performed on each aliquot using 10 second stimulation times, and the recovered doses were plotted against the applied doses (Fig. 8.4). The ratio of recovered dose to applied dose for the Luxel and quartzite rock slices was 1.02 and 0.97, respectively. These values are both exceptional, being within 3% of unity.

8.4 Pixel Size

The overall goal of LuCIDD is to develop dose maps, i.e., to stimulate multiple points on each surface. The size of the focused laser spots was found to be on the order of tens of microns, and one could therefore assume that the resolution of LuCIDD is on the same order. However, the actual area or volume from which luminescence originates is

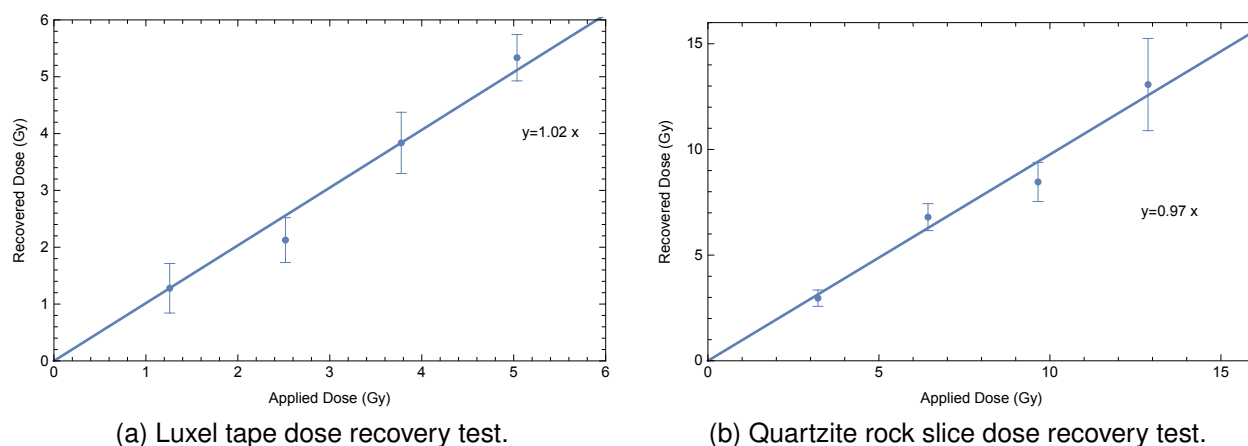


Figure 8.4: Dose recovery tests were performed on two materials, providing excellent agreement between the recovered dose and the laboratory applied dose.

likely larger. The laser light is scattered by the atoms in the mineral grains. Stimulated luminescence is emitted in all directions and might in turn stimulate more luminescence from defects that are outside the focal spot of the laser. The luminescence is refracted at grain boundaries. This behavior will depend on the mineral composition (opaque versus translucent) and the grain sizes of the rocks. To determine the effective resolution of LuCIDD the “radius of influence” or “effective bleaching radius” of a laser spot has to be measured.

Luxel dosimeter tape was used to determine an acceptable pixel size for developing dose maps. Five aliquots were irradiated with 11.61 Gy of X-ray dose and two points were measured on each aliquot with varying separation. Table 8.5 outlines the SAR procedure that has been modified to measure two points on a sample’s surface. This resulted in a recovered dose for both points relative to the initial applied dose. The recovered dose of the second stimulation point was divided by that of the first to obtain a relative dose value. Figure 8.5 shows a CCD image of the bleaching spot on the surface of the Luxel dosimeter tape at multiple separations.

Figure 8.6 shows the dose ratio of the two points plotted against the separation distance. It can be seen that the dose ratio is below unity for points that are separated less than 1.5 mm. This means that the stimulation of the first spot affected the second

Table 8.5: Steps of the SAR cycle for multiple measurement points on Luxel dosimeter tape.

Step	Action
1	Irradiate with a dose
2	Measure OSL at point 1 for 10s
3	Move to point 2 and Measure OSL for 10s
4	Irradiate with test dose
5	Measure OSL at point 1 for 10s
5	Move to point 2 and Measure OSL for 10s
7	Repeat steps 1-6 for each regeneration and test point

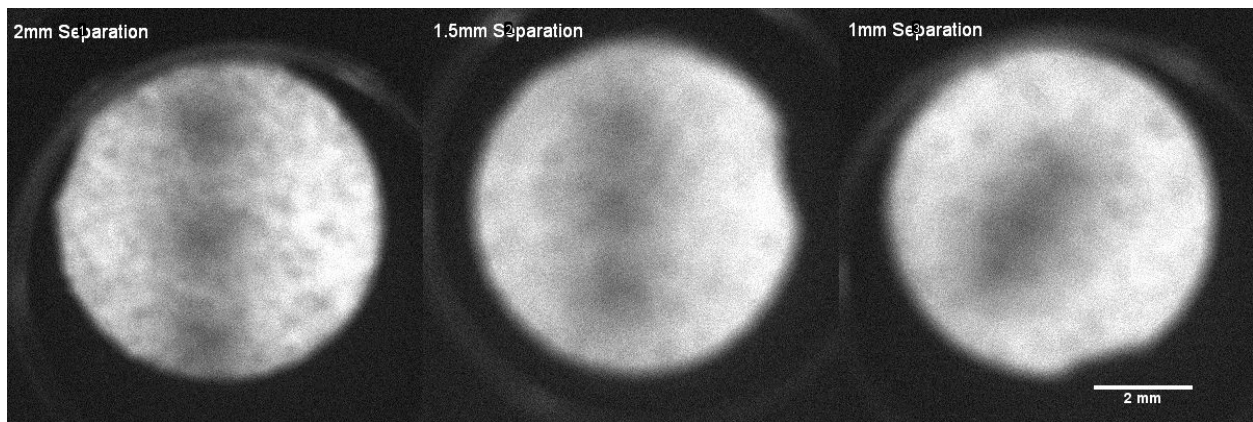


Figure 8.5: Images of the bleaching spot for 2 mm, 1.5 mm, and 1 mm separations. The spots on the 1 mm separation are barely resolvable, but the signal produced by the second spot doesn't allow for accurate equivalent dose measurements.

spot as well and partially bleached the luminescence. Due to the confocal setup, this luminescence does not reach the PMT and does not contribute to the signal of the first spot. However, when the second spot is measured, the signal is already partially depleted. While a separation of 1.5 mm has error bars overlapping with unity, a stimulation separation of 1.75 mm is suggested to improve precision.

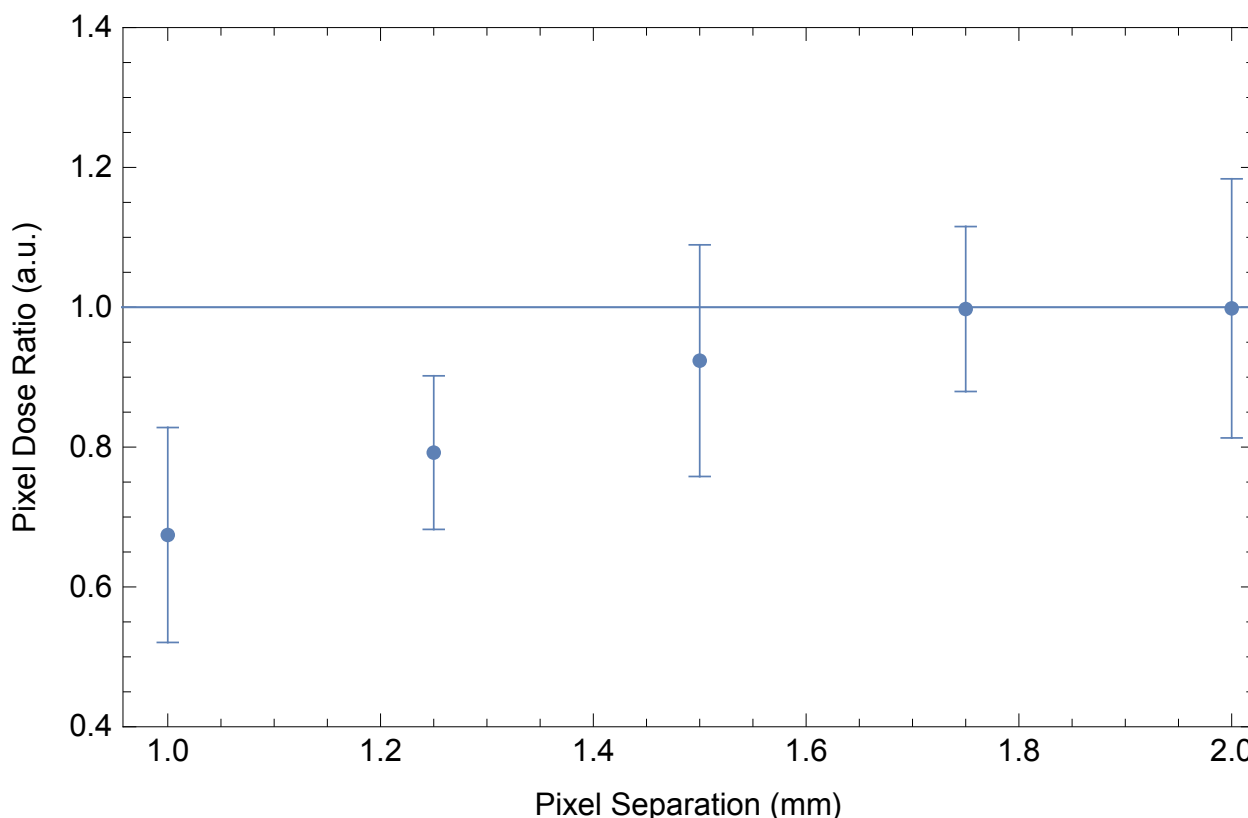
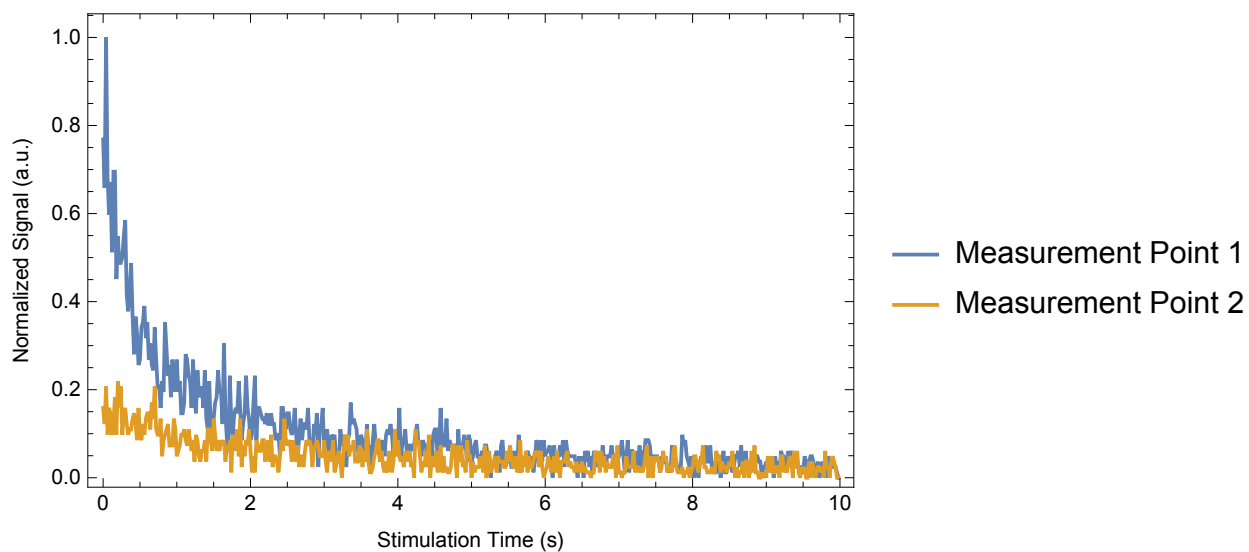
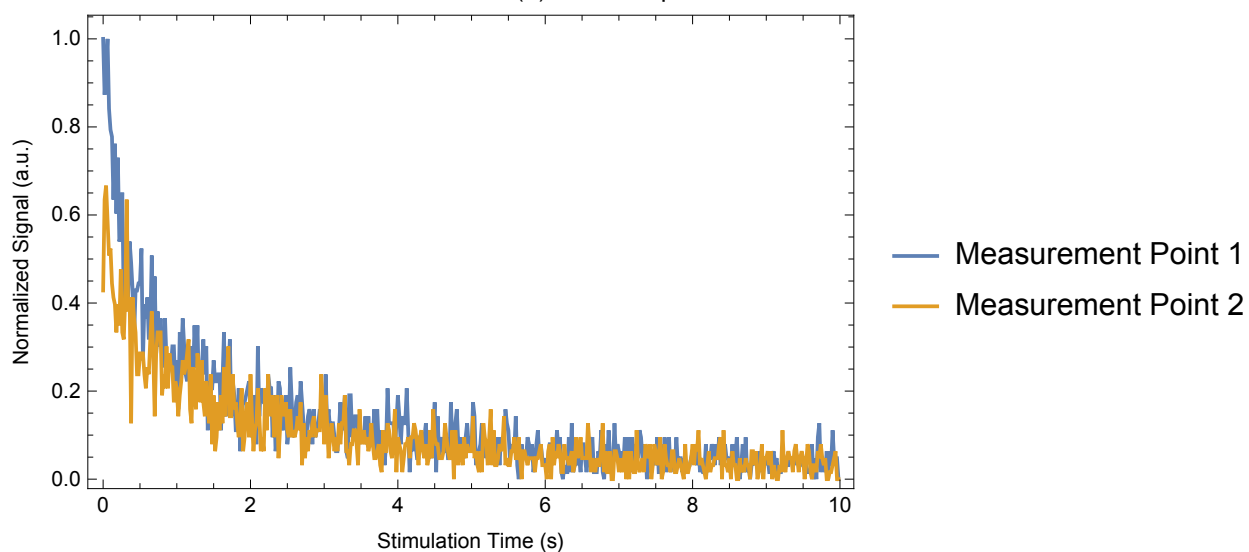


Figure 8.6: Pixel dose ratio versus the pixel separation. The pixel dose ratio improves with increasing separation until becoming unity at a 1.75 mm separation.

Figure 8.7 shows the natural OSL curves for the 1 mm and 2 mm separation measurements. The natural signal was reduced by a factor of five, but still retained enough signal to provide a dose that was 67% of the original value. It appears from figure 8.7a that even if the stimulation from the first point causes a reduced signal in the second point, a dose value might still be able to be found. This is confirmed in figure 8.7b with the 2 mm separation point that provided a unity dose ratio.



(a) 1 mm Separation



(b) 2 mm Separation

Figure 8.7: OSL curves of the measured natural signals for 1 mm and 2 mm separations. The OSL of each plot was normalized by the signal obtained from the first measured point. Natural signal of the second point is almost completely bleached for the 1 mm separation, while the 2 mm separation produces almost the same signal at the second point as it does with its first.

8.5 Discussion

In this chapter, the performance of LuCIDD in the SAR procedure was tested. It was shown that lasers may be focused on sample surfaces, but the shapes of the peaks are not ideal gaussians and they have multiple irregularities. This could be due to a misalignment of the beam path, resulting in either beams or components that are not perpendicular to the sample tray. If this is the case, then as the objective scans the z-axis the laser will drift from its initial (x, y) point to a different point with a different surface height and thus, focal length. This positional drift could result in elongated and multiple peaks being observed during the focusing scans.

It is also interesting to note that figure 8.2 shows that the shapes of the two green laser peaks show some similar irregularities. This is also true for the two green dichroic mirror peaks. This indicates that the irregularities could be laser- or mirror-dependent.

The precision of the autofocus could be lacking due to the large size of the confocal detection pinhole. LuCIDD currently makes use of a 250 μm detection pinhole which is about 13 Airy units relative to the IR laser, which has the largest Airy unit of the lasers. Reducing the pinhole to one Airy unit would increase the resolution of signal collection to 0.85 μm in the x-y plane and 3.0 μm along the z-axis. Note that this would only increase the resolution of the signal collection, and not the sample stimulation which has been found to be the major governor of pixel size. Lastly, decreasing the size of the detection pinhole would also decrease the signal received during stimulation, a quantity that is tied to the quality of the measurement and is already lacking for even the brightest samples.

Dose recovery tests carried out with LuCIDD performed well, with ratios of recovered doses to applied doses within 10% unity. Because of these passed tests, it can be confidently said that LuCIDD is capable of performing dosimetry measurements using optically stimulated luminescence; however, LuCIDD has only generated up to two equivalent dose points from a single sample and has yet to build an entire dose map.

LuCIDD's minimum acceptable stimulation time was observed to be 10 seconds for Luxel dosimeter tape, providing relative standard errors of 7.41% for single dose recovery points. Applying this stimulation time resulted in a minimum resolvable pixel size of 1.75 mm. This pixel size is much larger than the 75 μm resolution applied by others with laser stimulation (Greilich, Glasmacher, and Wagner 2005). It is possible that reducing the stimulation time could result in smaller bleached areas around the stimulation point and thus, better pixel resolution, but would increase the uncertainty in any measurements made due to the decrease in signal collection. Further conclusions and suggestions for future research are discussed in the next chapter.

Chapter 9

Conclusions and Future Directions

The goal of this dissertation was to develop an instrument suitable for producing spatially-resolved luminescence dose maps from whole rock surfaces and other dosimetric materials. The basic requirements for spatially resolved luminescence measurements were discussed in chapters 3 and 4. Components suitable for producing equivalent dose maps on sample surfaces were characterized and those components were integrated into a single luminescence reader called LuCIDD (chapters 5-7). Finally, the viability of LuCIDD's application to dose mapping was evaluated in chapter 8. This chapter provides a discussion of the work performed in this dissertation and suggests possible directions for future research.

9.1 Dissertation Summary

Similar to conventional and commercial readers, LuCIDD is equipped with all of the necessary components for luminescence dosimetry, including facilities for stimulating luminescence with light and heat, a light detector, and a calibration source. To allow for dose mapping, LuCIDD's design was based on a confocal microscope. One of three different lasers can be focused onto the sample surface to stimulate luminescence from

“pixels”. The confocal design of the detection system allows rejection of the sub-surface luminescence, which might originate from depths that had not been sufficiently reset by sunlight. For the calibration source, an X-ray emitter was selected instead of the more common beta source.

The focal spot size, power density, and penetration depth of the stimulating lasers were characterized in chapter 6. The focal spot sizes that were acquired were very small at $<64.5\text{ }\mu\text{m}$, while their power densities were $>10^2\text{ W/cm}^2$. The penetration depths of the lasers were found to be less than 1 mm.

Chapter 7 characterized the X-ray source for energy, beam uniformity, dose linearity with current, dose rate, stability, and penetration depth. A $50\text{ }\mu\text{m}$ aluminum filter was chosen to be applied with an 18 mm collimator to the X-ray source to harden the lower energy bremsstrahlung emitted by the source without losing beam intensity. The spatial variation of the X-ray beam was less than 5% of the mean applied dose within an 18 mm sample area. The source showed excellent linearity with current and was stable over short or long lengths of time. The dose rate of the X-ray source was found to be comparable to the Risø’s beta source for all natural samples and was 69% higher for the Luxel. Lastly, the penetration depth of the X-ray source was measured to be less than 0.5 mm, which is between the minimum and maximum penetration depths found for the lasers.

Finally, in chapter 8, the minimum stimulation time and pixel size were determined, and dose recovery tests were performed. Dose recovery tests on individual points on a sample exceeded expectations by remaining within 3% of unity for both Luxel dosimeter tape and quartzite rock slices, indicating that LuCIDD is well suited for accurate dose recovery. A stimulation time of 10 seconds was chosen and led to a pixel size of 1.75 mm to limit the error in equivalent dose measurements, but more work is required to measure a dose map.

9.2 Discussion

The small focal spot sizes of the lasers did not translate into accordingly small pixel sizes. The pixel sizes obtained in this research were determined to be 1.75 mm while others have achieved resolutions at less than 5% of this (Greilich, Glasmacher, and Wagner 2005). They had obtained this resolution using a low power (24 mW/cm^2) green, 532 nm laser and long stimulation times (180 seconds). It has been discussed in chapter 6 and chapter 8 that LuCIDD's high stimulating power coupled with its 10 second stimulation times could result in excessively large bleaching radii, therefore increasing pixel size. The 10 second stimulation time was selected to reduce the error in measurements, but a shorter time could be selected to balance resolution with error. The tests performed here were carried out with Luxel, which consists of small single grains in a plastic matrix, and further tests are necessary to determine LuCIDD's pixel size when a solid material with larger grains, such as a rock surface, is used.

The lasers' penetration depths were small ($<1 \text{ mm}$), but this does not necessarily equate to the depth at which they bleach OSL traps; the question still remains from what depth the OSL signal originates. OSL intensity decreases with stimulation intensity and, like the laser light, the OSL signal is also attenuated as it passes through the rock. If the laser bleaching depth is truly less than 1 mm, then depth resolution may not be required by LuCIDD at all. If this were the case, LuCIDD's $250 \text{ }\mu\text{m}$ detection pinhole could be removed entirely for signal collection. This would allow the PMT to record more signal and could reduce the stimulation time required to achieve an acceptable amount of signal while reducing the laser's bleaching radius and allowing finer resolution measurements.

However, X-rays are attenuated in the material as well, leading to a dose gradient with depth. While dose-recovery tests from individual points have been successful, the compounding effects of dose gradients, stimulation intensity gradients and spatial origin of the OSL require further study.

9.3 Future Directions

Much progress has been made in this dissertation, but there are still many steps necessary to achieve the goal of dose mapping with LuCIDD. The list of suggestions for future work could be several pages long. They include improvements to the instrument's setup, further study of laser and X-ray depth profiles, quantifying spatial effects of laser stimulation. The items listed here pertain only to the most important tasks.

There are two main directions for future research. First, LuCIDD could be tested in its current state to provide dose mapping of non-uniform radiation fields with resolutions of about 1.75 mm x 1.75 mm. This resolution would be less suited to dating rocks or sediments with small grain sizes ($<250\text{ }\mu\text{m}$) but could be used to characterize non-uniform dose fields in medicine or space applications.

For the second direction, tests could be performed to determine if a new iteration of LuCIDD could be developed without the use of a confocal detection pinhole. The tests would include determining the true bleaching depth of the lasers at different power levels and stimulation times, as well as performing additional characterizations on the axial component of the X-ray source's spatial variation.

During the course of this research, LuCIDD was constructed, its components were successfully tested, and measurements on individual points provide a proof-of-concept for LuCIDD's ability to accurately obtain equivalent dose information. In conclusion, the work carried out in this dissertation equated to a large leap toward measuring dose maps with LuCIDD, and the future prospects of LuCIDD's application to dose mapping remain healthy.

Appendix A

Software Capabilities of the Risø

TL/OSL Reader

The software used to control and command the Risø, and its measurements are the Sequence Editor, the Viewer, the Analyst, and the Control programs. Due to the modular nature of the Sequence Editor program, users can build a combination of measurements and treatments from a list of operations including irradiation, preheat, TL, OSL, and more. These combinations may be applied to multiple carousel positions, or a different set of operations may be performed on any desired position. For instance, one may decide to perform a dose recovery test on four aliquots of one sample, a complete SAR on twelve aliquots of another sample, apply irradiations to four more aliquots of the previous sample, and leave the last four carousel slots unused. These sequences may be saved for later use and the Sequence Editor provides a live display of TL and OSL signals on screen while the sequence is being performed (Fig. A.1). The recorded data is saved and is accessible to both the Viewer and the Analyst.

The Viewer allows the viewing and printing of recorded curves as well as the exportation of the data into other usable file types. The Analyst allows all of this and enables the analysis of the data for luminescence dating and dosimetry purposes. In essence, the

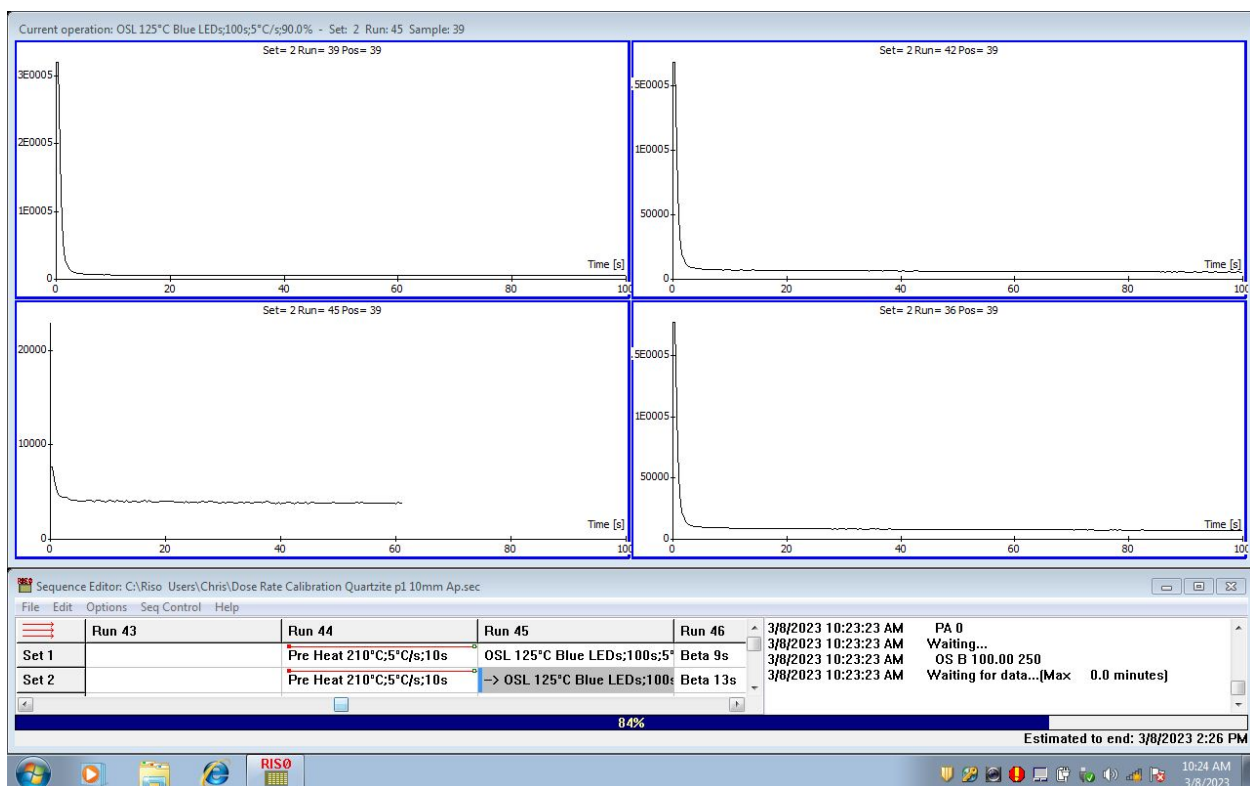
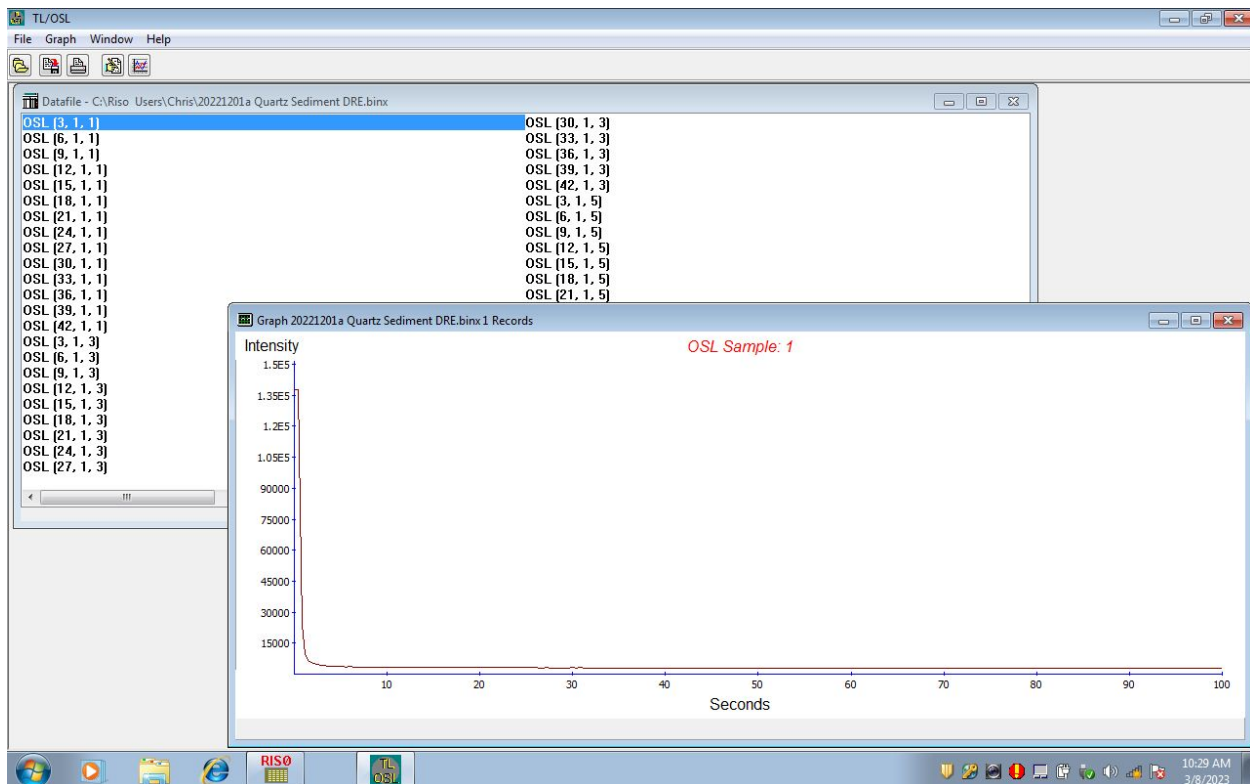
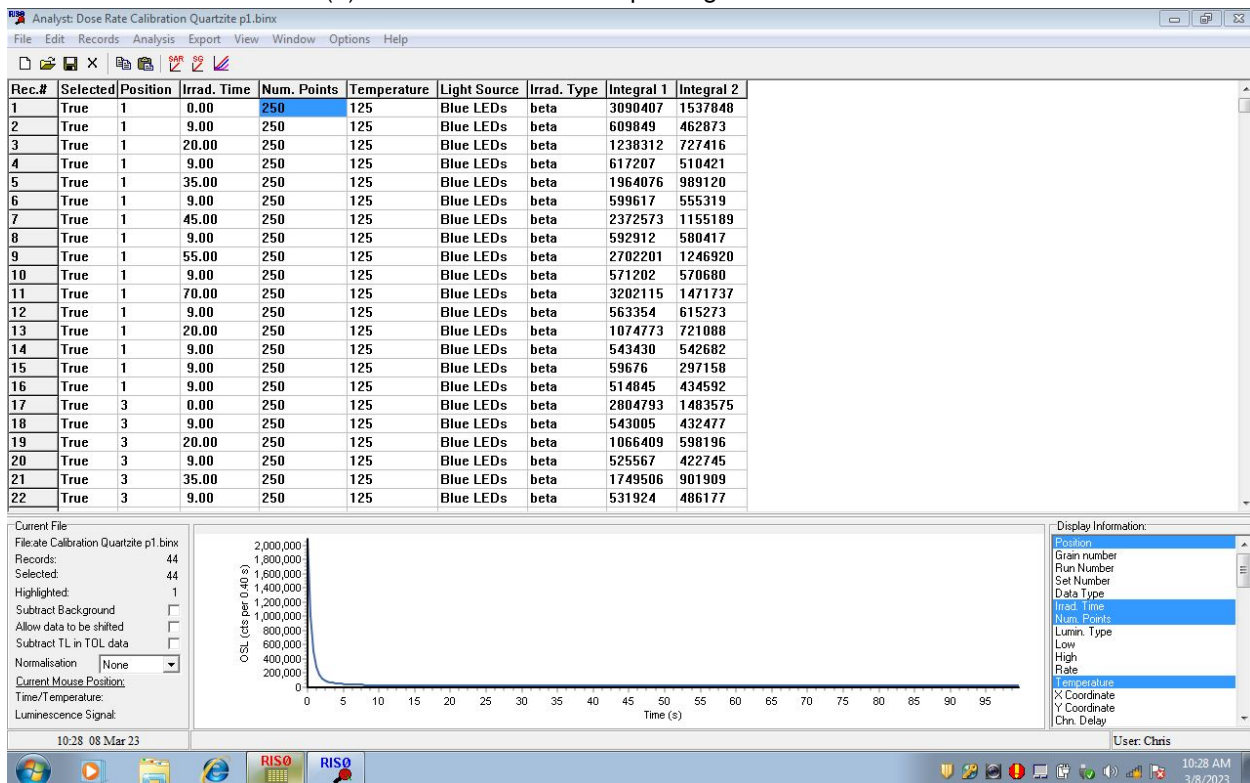


Figure A.1: Screenshot of the Sequence Editor and the live, on-screen display of the OSL signal. A preheat at 210°C and an OSL measurement with blue LEDs is being performed.

Analyst is a more robust version of the Viewer application (Fig. A.2). The Control program allows the testing of each individual action that may take place during sample loading and measurement. Note that the “Blue Diodes On” option will activate the green LEDs if that modular array is currently attached to the Risø.



(a) The Viewer allows the printing of visualized data.



(b) The Analyst is used to view and analyze data.

Figure A.2: The Viewer and the Analyst programs are used to visualize and quickly analyze datasets obtained from the Risø.

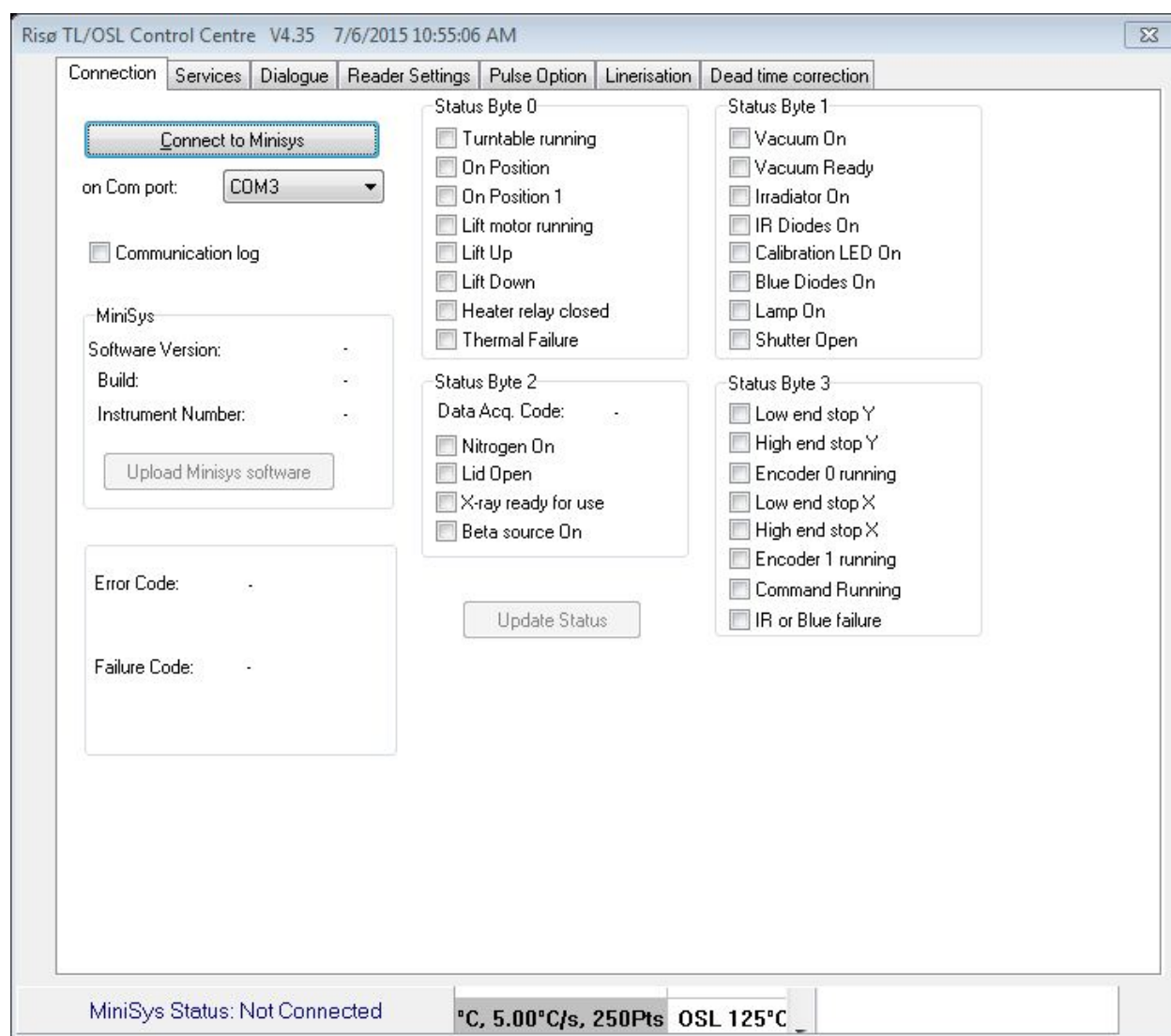


Figure A.3: The control program allows manual direction of the Risø reader. Using the Control program, the lid, lift, heater, nitrogen, LEDs, radiation source, PMT, and other components may be manually tested and operated.

Appendix B

Verification of the SAR Protocol

At the end of the SAR procedure, two test points are added that are used to determine if a sample can reliably recover a dose. These two test points are called the recycling ratio and the recuperation. The recycling ratio is used to determine the effectiveness of the test dose sensitivity correction by repeating the irradiation and measurement of the first regeneration point. The relative signal from the recycling ratio test point is divided by the relative signal from the first regeneration point resulting in a value of unity for a well-behaved sample. An aliquot is rejected if the recycling ratio provides values outside of 1.0 ± 0.1 .

The recuperation test is used to determine the effects of thermal treatment on the luminescence signal. A sample that is properly reset should produce little to no observable OSL signal, but thermal treatment (in the form of the preheat step) has been shown to create residual signals in samples, post-OSL (Murray and Wintle 2003). This effect is called thermal transfer. Thermal transfer causes the release and re-trapping of electrons from shallow, non-radiative traps into deeper, optically sensitive traps. These traps are then evacuated during the stimulation step, inflating the luminescence signal. To determine if an aliquot is affected by thermal transfer, the recuperation test is applied. This test point consists of a $D_i = 0$ Gy SAR cycle where the signal obtained from the point is compared

to that of the natural. An aliquot is rejected if the recuperation signal is greater than 5.0% of the natural signal.

B.1 Determination of the Preheat Temperature

The preheat step is important for emptying traps that would be empty in nature but become filled during laboratory irradiations. The ideal preheat temperature is determined by taking subsets of aliquots and running the SAR procedure at different temperatures for each subset. The determined natural doses are plotted against their preheat temperatures, resulting in a plateau of doses (Fig. B.1). The doses tend to decrease at lower temperatures and increase at higher temperatures. The preheat temperature is chosen from the center of the plateau. In the example in the figure, the plateau ranges from 180 to 200°C and a preheat temperature of 190°C would be selected.

B.2 Testing the SAR Procedure

A more substantial test of the overall SAR procedure is the dose recovery test, shown in figure B.2. This test is used to determine the ability of the sample to accept SAR as a dating protocol, as well as testing the effect of heat treatment on the luminescence signal. Testing this effect is important because during the SAR, the natural signal is measured *before* any laboratory heat treatment is applied, but the regenerative signals are all measured *after* heating. To perform the dose recovery test, multiple subsets of aliquots are optically reset without exposure to heat. The subsets are each given a different laboratory dose to act as their “natural” dose. Recovery of the laboratory-induced natural doses are attempted using the SAR procedure. The recovered dose is divided by the known applied dose and should be equal to unity. The entire sample may be rejected if the dose recovery test provides ratios outside of 1.0 ± 0.1 .

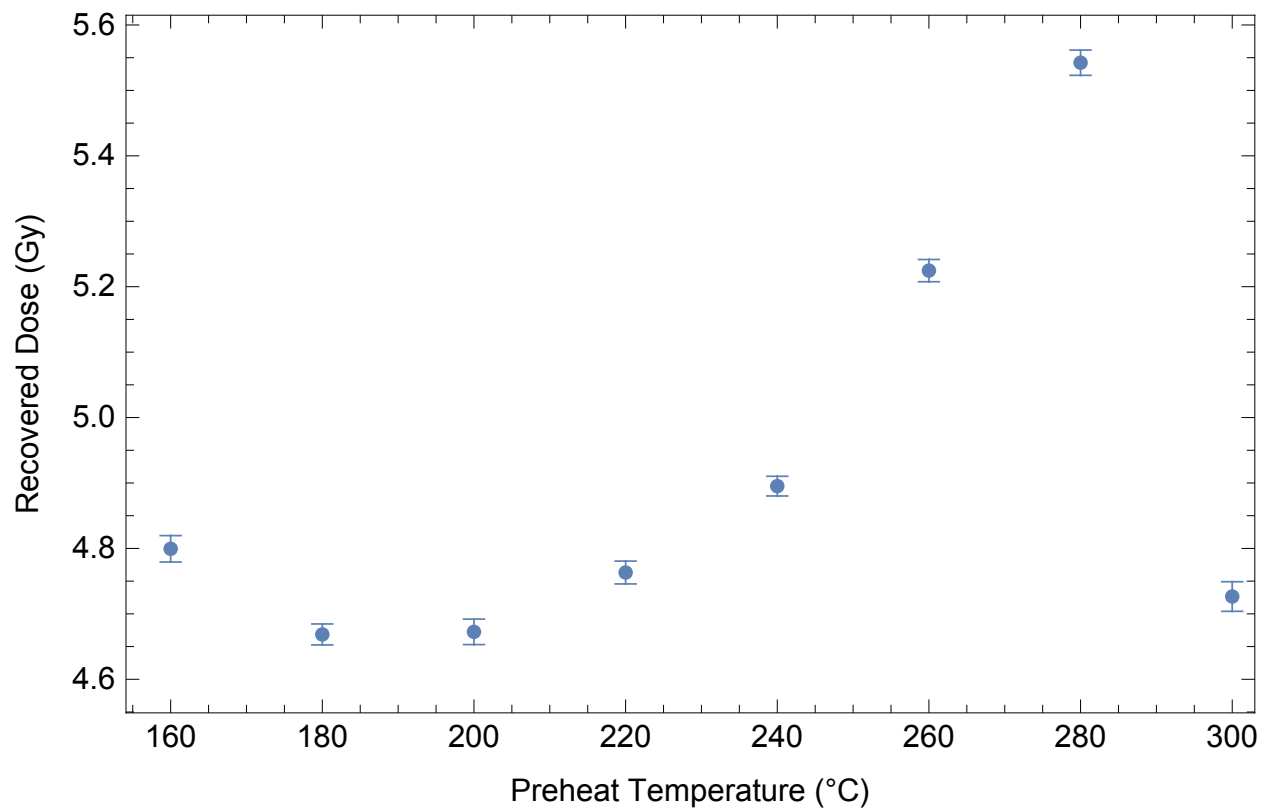


Figure B.1: The SAR procedure is applied to the sample at varying preheat temperatures to determine the effect of the preheat temperature on the luminescence signal. The recovered dose increases drastically with increasing temperature.

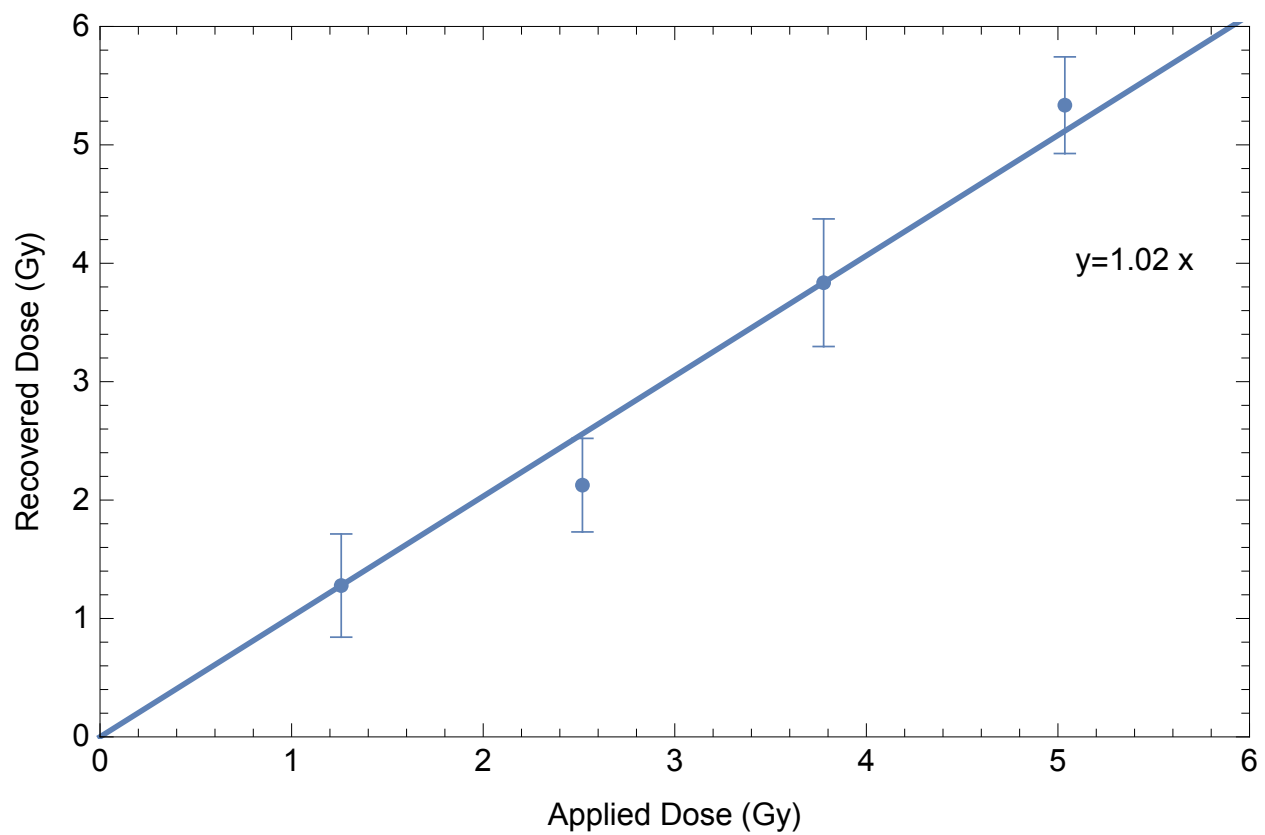


Figure B.2: Example of a dose recovery test. The recovered doses are plotted against the applied doses and the slope of the fit line should be one for a perfect test.

Appendix C

Theory of X-Ray Tubes

C.1 Hot-Cathode X-Ray Tube Construction

The spatial uniformity and diameter of an X-Ray tube's output beam is a direct result of the construction of the tube. Hot-cathode X-Ray tubes consist of a cathode (filament) and an anode (target) with an applied potential difference inside of a vacuum chamber (C.1). The filament is heated by an applied current and electrons are excited and ejected from the cathode through a process called thermionic emission. These ejected electrons are accelerated toward the anode due to the external potential applied across the tube. When the high-energy thermoelectrons collide with electrons within the target, the target electrons are ionized and characteristic X-Rays are released with energies dependent upon the material of the target (Knoll 2010). Other X-Rays are emitted in a continuous spectrum through a process called Bremsstrahlung; this process occurs as electrons become accelerated by nuclei they pass, releasing energy in the form of X-Ray photons.

X-Rays emitted from the anode tend to be directed away at right angles relative to a low-energy electron beam but are produced in lesser quantities at other angles (Johns and Cunningham 1983). This results in a higher photon intensity at right angles to the electron beam than any other angle. That is to say, the spatial uniformity of the X-Ray

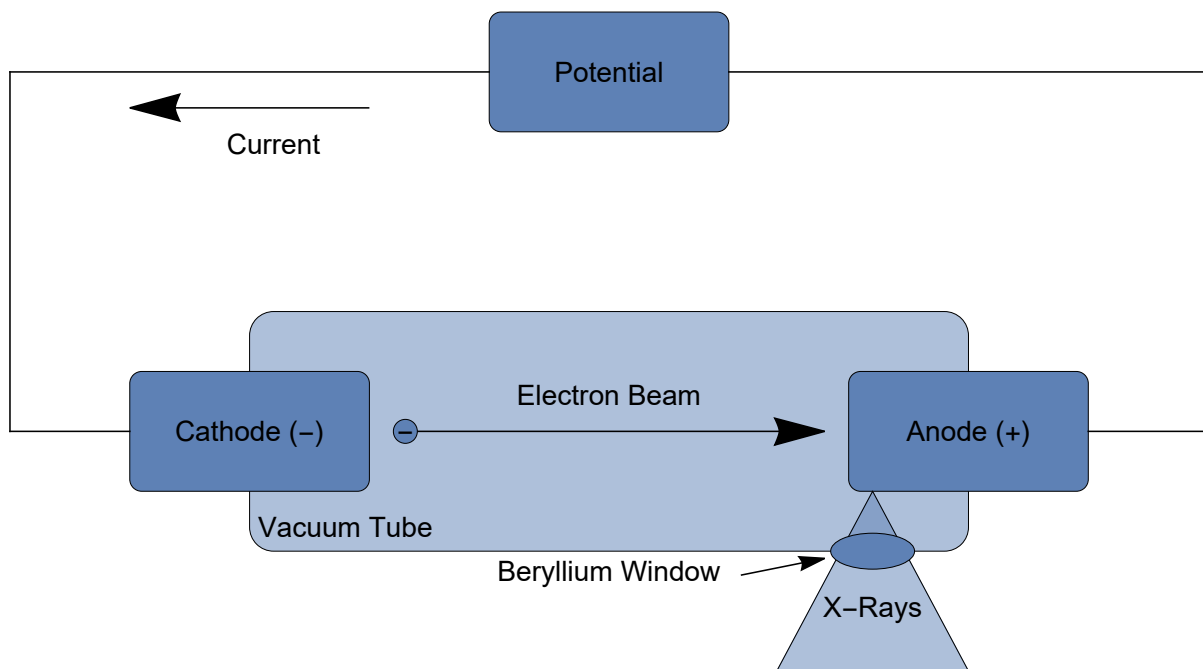


Figure C.1: An X-Ray tube consists of a negative cathode and a positive anode. The cathode is heated and emits electrons which are accelerated towards the anode. X-Rays are produced as the electrons impact the anode and may escape through the beryllium window.

beam is dependent upon the relative probability of the emission angle of the emitted photons. X-Rays that exit the tube through the beryllium window are applied as dose to samples. These escaping X-Rays form a cone with an apex angle relative to the width of the beryllium window and the target-to-window distance. Thus, the emission angle of the X-Ray tube is the primary driver for the diameter of its beam.

C.2 Thermionic Emission of Electrons

Introduced in the previous subsection, thermionic emission occurs when the tube's filament is heated by an applied current. Unbound electrons are free to randomly move inside a metal with a velocity distribution that is dependent upon the temperature T of the metal. As the filament of the X-Ray tube becomes heated by the applied current, electrons are “boiled” off the surface of the filament (Rosa and Ordóñez 2022). The minimum amount of energy required to excite and eject an electron from the filament is called the work function and is the product of the electron charge q and the potential ϕ of the system. This work function is related to the thermionically emitted current I :

$$I = \frac{4\pi m q k^2}{h^3} T^2 e^{\left(-\frac{q\phi}{kT}\right)} = A_{th} T^2 e^{\left(-\frac{q\phi}{kT}\right)} \quad (\text{C.1})$$

with m as the mass of the electron, k as Boltzmann's constant, h as Planck's constant, and A_{th} as the thermionic constant in units of A/m²K². The potential (and thus the work function) is a “well-behaved” function of temperature, and may be written as a sum of terms:

$$\phi = \phi_0 + a_1 T + a_2 T^2 + \dots \quad (\text{C.2})$$

Here, ϕ_0 is the work function potential at absolute zero. Taking only up to the linear term gives us a more useful equation for the thermionic current:

$$I = A_{th} T^2 e^{\left(-\frac{q\phi_0}{kT}\right)} e^{\left(-\frac{qa_1}{k}\right)} = AT^2 e^{\left(-\frac{q\phi_0}{kT}\right)} \quad (\text{C.3})$$

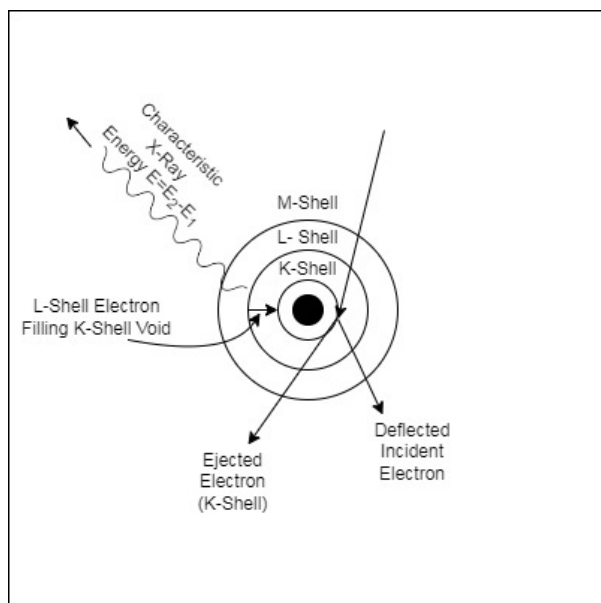
with $A = A_{th} e^{\left(-\frac{qa_1}{k}\right)}$ as the emission constant.

C.3 Interactions of Electrons in Matter

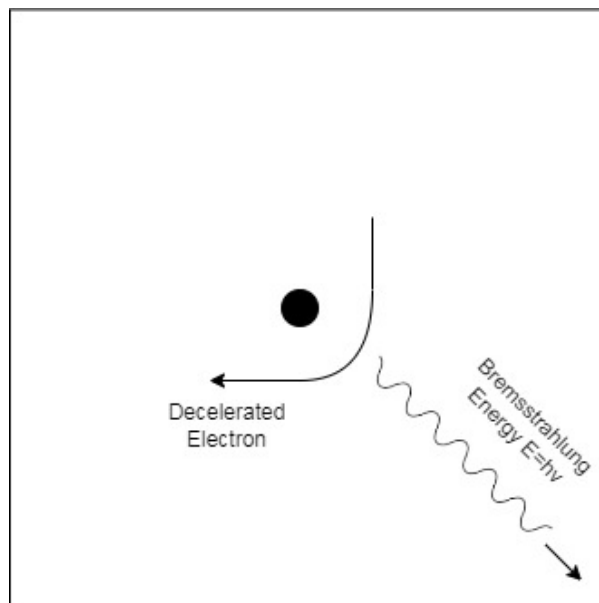
The electrons that make up the thermionic current are accelerated through vacuum until impacting the target anode. As the electrons travel throughout the target, they lose energy due to ionization and radiative interactions – some of which cause a release in energy in the form of X-Rays.

Ionization interactions occur when thermionic electrons transfer enough energy to atomic electrons through collisions to liberate them from the atom, creating a vacancy in their shell. As a higher-shell electron drops down to fill the newly created void, it will release its stored energy as a photon of energy $h\nu$ (C.2a). Photons emitted in this way are called characteristic X-Rays, whose energies are dependent upon the anode material. Ionization losses that end up in X-Ray production make up $\sim 1\%$ of the energy loss due to ionization while the rest of the energy is released as heat. Because of this, X-Ray sources are quite inefficient at producing X-Rays and generally require active cooling to prevent overheating.

Radiational losses are barely more efficient X-Ray producers at $\sim 1\%$ of the *total* energy loss creating X-Rays. As electrons travel close to positively-charged nuclei within the target, they are dragged into a partial orbit while slowing down. The energy loss from this deceleration is radiated as a photon in the X-Ray range (C.2b). X-Rays produced in this way are suitably named Bremsstrahlung, which is German for “braking radiation.” Because the effect of the deceleration of the incident electron is not quantized, the produced X-Rays are continuous in energy.



(a) A high energy incident electron ionizes a K-shell electron in the anode material. Characteristic X-Rays are produced when an electron from a higher shell fills the K-shell vacancy.



(b) As an incident electron is decelerated by positively-charged nuclei it releases energy in the form of Bremsstrahlung.

Figure C.2: Schematic of ionization (a) and radiative (b) losses in the anode of an X-Ray tube.

C.3.1 Ionization Collisions and Characteristic X-Rays

Characteristic X-Rays occur when an electron from a higher shell fills a vacancy in a lower shell, usually the K-shell. As it does, the electron releases energy as an X-Ray with energy equal to the difference in the binding energies between the two shells: $E = E_2 - E_1 = h\nu$. Due to the quantized nature of binding energies, the X-Rays produced are of a wavelength that is specific to the material being bombarded, hence, characteristic X-Rays. The processes that occur in the anode to eject these K-shell electrons are called “hard” collisions. These are direct collisions between the electrons excited from the cathode and the electrons housed in the atoms of the target material. Not all ionization losses produce X-Rays; “soft” collisions may occur between incident electrons and whole atoms that cause the electron to impart energy to the atoms through Coulomb forces. This increase in energy could cause the atom to eject a valence electron which could have enough energy to create its own track, known as a δ -ray.

The rate of energy loss per unit of path length per unit of medium density is called the mass stopping power ($S = 1/\rho \frac{dE}{dx}$) and is the force responsible for slowing down charged particles in a medium. For electrons traveling through a target anode, the mass stopping power is a combination of two terms: the ionization mass stopping power (S_{ion}) and the radiative mass stopping power (S_{rad}). The ionization mass stopping power can be further separated into soft and hard collision mass stopping powers,

$$S_{ion} = \frac{dE_s}{\rho dx} + \frac{dE_h}{\rho dx} \quad (C.4)$$

with the coefficients s and h being soft and hard, respectively. Expanding the right-hand side:

$$S_{ion} = \int_{E'_{min}}^H E' Q_s dE' + \int_H^{E'_{max}} E' Q_h dE' \quad (C.5)$$

Here, Q_s and Q_h are the mass collision coefficients for soft and hard collisions, E' is the energy transferred from the incident particle to the atom or electron in the medium, H is an energy boundary between the soft and hard collisions, E'_{max} and E'_{min} are the maximum and minimum transferable energies, and are related by:

$$\frac{E'_{max}}{E'_{min}} \approx \left(\frac{2m_0 c^2 \beta^2}{I} \right)^2 \quad (C.6)$$

with $\beta = v/c$, $m_0 c^2$ as the rest mass of an electron, and I as the mean excitation energy of the atom in the medium. The first term of the mass stopping power is given by the Bethe formula (Evans 1955):

$$\frac{dE_s}{\rho dx} = \int_{E'_{min}}^H E' Q_s dE' = \frac{2C m_0 c^2 z^2}{\beta^2} \left[\ln \left(\frac{2m_0 c^2 \beta^2 H}{I^2 (1 - \beta^2)} \right) - \beta^2 \right] \quad (C.7)$$

The constant $C = \pi r_0^2 Z N / A$ is the product of the classical electron radius r_0 and the number of electrons per gram of the medium $Z \times N / A$. The term outside the brackets is

generally simplified as $k = 2Cm_0c^2z^2/\beta^2$:

$$\frac{dE_s}{\rho dx} = k \left[\ln \left(\frac{2m_0c^2\beta^2 H}{I^2(1-\beta^2)} \right) - \beta^2 \right] \quad (\text{C.8})$$

For relativistic particles, we may rewrite β using the kinetic energy of the particle:

$$\beta = \left[1 - \left(\frac{1}{E/m_0c^2 + 1} \right)^2 \right]^{1/2} \quad (\text{C.9})$$

Such that $\frac{\beta^2}{1-\beta^2} = \frac{E(E+2m_0c^2)}{(m_0c^2)^2}$. The soft collision term then becomes:

$$\frac{dE_s}{\rho dx} = k \left[\ln \left(\frac{2HE(E+2m_0c^2)}{m_0c^2I^2} \right) - \beta^2 \right] \quad (\text{C.10})$$

While this equation is useful for all charged particles, that of hard collisions differs depending on the particle. Determining the total mass stopping power for the collisions of light particles like electrons requires quantum mechanical considerations and will not be discussed in detail here. The total collision mass stopping power for electrons in a medium is:

$$\frac{dE_{total}}{\rho dx} = k \left[\ln \left(\frac{E^2(E+2m_0c^2)}{2m_0c^2I^2} \right) + \frac{E^2/8 - (2E+m_0c^2)m_0c^2 \ln 2}{(E+m_0c^2)^2} + 1 - \beta^2 - \frac{2C}{Z} - \delta \right] \quad (\text{C.11})$$

Here, $-2C/Z$ is a term used to correct for decreasing atomic shell electron participation at lower particle velocities. As the incident electron's velocity decreases below the velocities of K-shell electrons in the medium, the stopping power contribution from that shell goes to zero, effectively decreasing the value of I which should then ignore the K-shell contribution. The δ term corrects the mass stopping power for the density of the material. In gases, collisions are assumed to be independent of one another. However, in a dense medium, there are contributions of nearby atoms and electrons other than those directly involved in the collision, actually *decreasing* the energy loss in the collision and

thus, slightly decreasing the mass stopping power. This term is negligible for energies less than the rest mass energy of the incident particles (0.511 MeV for electrons.)

C.3.2 Radiative Losses and Bremsstrahlung

Bremsstrahlung occurs when electrons radiate energy during non-ionizing collisions. The nature of the electron's energy loss allows bremsstrahlung to form a continuous energy spectrum with the highest intensities found near zero energy and decreasing as the Bremsstrahlung energy increases to the maximum allowed energy, equal to the accelerating potential of the X-Ray source.

The mass stopping power of radiative losses is a result the theory behind the Bethe formula (Evans 1955):

$$\frac{dE_r}{\rho dx} = \sigma_0 \frac{N_A Z^2}{A} (E + m_0 c^2) \bar{B}_r \quad (\text{C.12})$$

Where the subscript r denotes radiative losses. The constant $\sigma_0 = 1/137(e^2/m_0 c^2)^2 = 5.80 \times 10^{-28} \text{ cm}^2/\text{atom}$, N , Z , and A are the neutron, atomic, and mass numbers respectively, and $\bar{B}_r$ is a function of Z and E . The mass stopping power for a silver anode has been plotted in figure C.3. The ionization mass stopping power dominates the total stopping power at low energies, where our X-Ray source operates but this does not negate the contributions of bremsstrahlung.

The maximum Bremsstrahlung energy may be manipulated by varying the source potential, while the intensity of X-Ray production may be controlled by varying the source current. In general, Bremsstrahlung reduces the average energy of the X-Ray output, reducing its penetrating power.

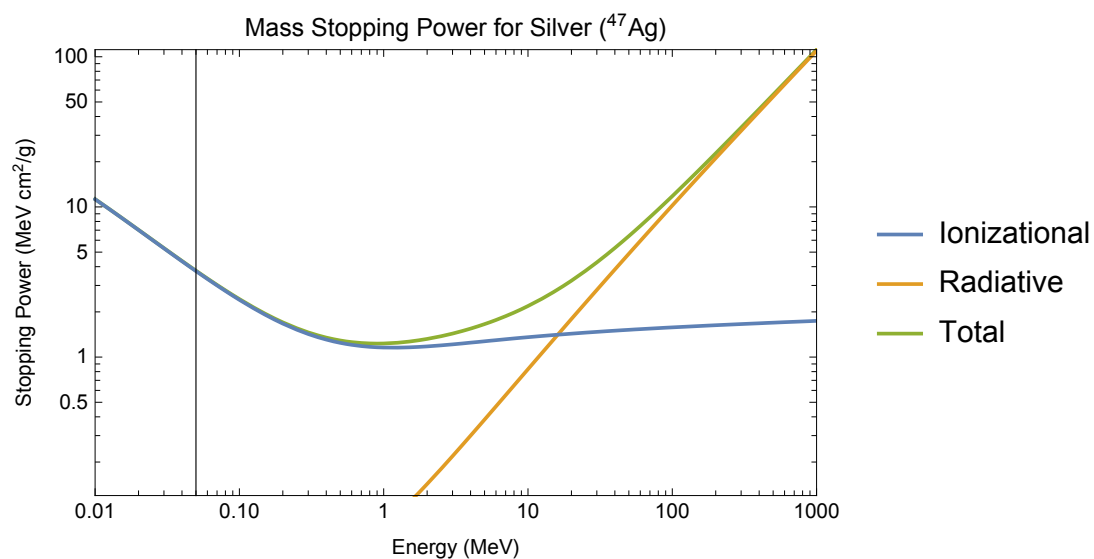


Figure C.3: Mass Stopping Power for Silver. The vertical black line shows the operating energy of LuCIDD's selected X-Ray source. This figure was created using data from the NIST ESTAR database. (Standards and Technology n.d.)

Appendix D

LuCIDD Part Numbers

Table D.1: Specifications of the lasers and companion components used in LuCIDD's stimulation unit.

Part	Wavelength (nm)	Power (mW)	Manufacturer	Part Number
IR Laser	852	100	CrystaLaser®	DL852-100
IR Laser Power Supply	-	-	CrystaLaser®	CL-2005
Red Laser	637	50	Coherent Cube®	1069415
Red Laser Power Supply	-	-	Coherent Cube®	1039966
Green Laser	532	100	MeshTel®	GSF32-100PS
Green Laser Power Supply	-	-	Sintech Optronics	VD-III A
Blue Laser	473	100	CrystaLaser®	CL472-100
Blue Laser Power Supply	-	-	CrystaLaser®	CL-2005
Laser Shutters (x4)	-	-	ThorLabs®	SH05
Laser Shutter Controller (x4)	-	-	ThorLabs®	SC10

Table D.2: Specifications of the geometrical optics used in LuCIDD's stimulation unit.

Part	Focal Length (mm)	Passing Wavelength (nm)	Manufacturer	Part Number
IR Beam Expander Objective Lens	-6.0	-	ThorLabs®	LC2969-B
IR Beam Expander Image Lens	+35.0	-	ThorLabs®	AC254-35-B
Red Beam Expander Objective Lens	-6.0	-	ThorLabs®	LC2969
Red Beam Expander Image Lens	+60.0	-	ThorLabs®	AC254-60-A
Green Beam Expander Objective Lens	-6.0	-	ThorLabs®	LC2969
Green Beam Expander Image Lens	+50.0	-	ThorLabs®	LA1131
Blue Beam Expander Objective Lens	-6.0	-	ThorLabs®	LC2969
Blue Beam Expander Image Lens	+40.0	-	ThorLabs®	AC254-40-A
Planar Mirror (x8)	-	-	ThorLabs®	PF10-03-P01-10
Red Dichroic Mirror	-	>720	Semrock	FF735-Di01
Green Dichroic Mirror	-	>570	Semrock	FF580-Di01
Blue Dichroic Mirror	-	>500	Semrock	FF506-Di03

Table D.3: Specifications of the components used in LuCIDD's confocal unit.

Part	Focal Length (mm)	Passing Wavelength (nm)	Manufacturer	Part Number
Beam Splitter	-	400-700 (70% Transmitting)	Edmund Optics®	30R/70T VIS
Green Dichroic Mirror	-	390-500	Chroma	T 500 SPXR
Blue Dichroic Mirror	-	300-400	Materion	N/A (Custom)
Reflective Microscope Objective	25.0	-	Edmund Optics®	50105-02
Pinhole Assembly Objective Lens	35.0	-	ThorLabs®	LA4052-UV
Pinhole Assembly Image Lens	35.0	-	ThorLabs®	LA4052-UV
Confocal Pinhole (250 μ m)	-	-	Edmund Optics®	N/A
Photomultiplier Tube	-	-	Hamamatsu®	H11890-110
UG-11	-	254-405	Schott	UG-11
330 WB80	-	250-410	Omega	330 WB80

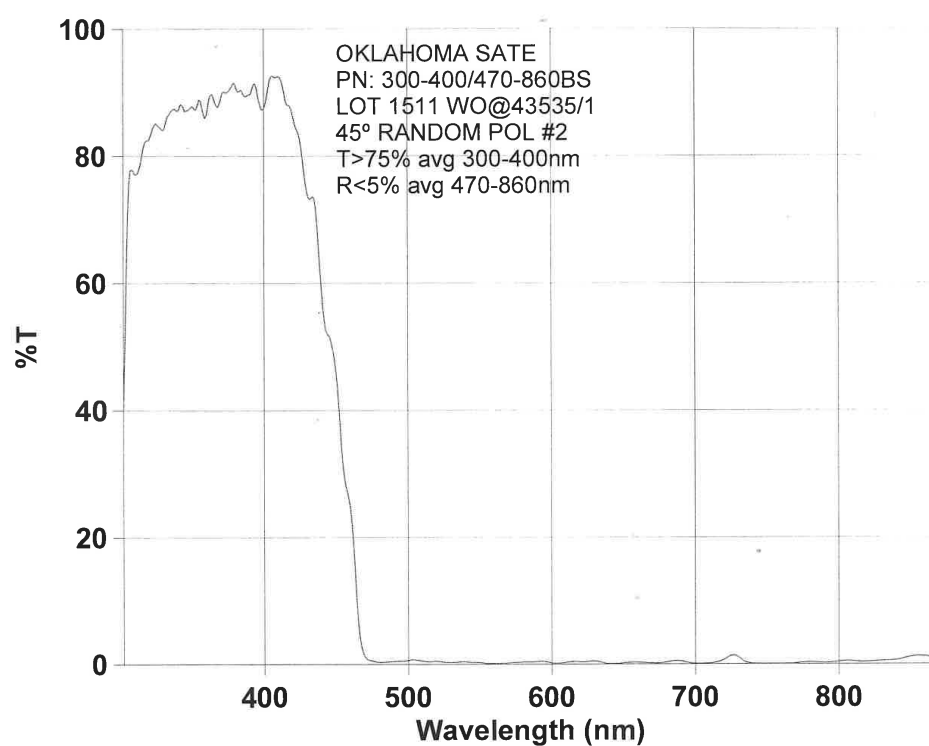


Figure D.1: Scan of LuCIDD's custom blue dichroic detection mirror datasheet.

Appendix E

Instrumentation and Method Details for X-Ray Calibrations

E.1 CdTe X-Ray Detector

An AmpTek XR-100-CdTe X-ray detector was used to determine the effect of filtration on LuCIDD's X-ray energy spectrum relative to multiple filters. The X-ray detector was calibrated using multiple survey sources. Once calibrated, the detector was used to measure the energy spectrum of LuCIDD's X-ray source when filtered and unfiltered. This subsection describes the methods used to calibrate the X-ray detector to allow measurement of the energy spectrum of the X-ray tube.

E.1.1 Instrumentation

Spectra were observed using an AmpTek XR-100-CdTe X-ray detector and were measured live by AmpTek's DPPMCA Display and Acquisition software. This software allows full control of hardware features like pulse shaping, threshold determination, and detector temperature. The X-ray detector was outfitted with a 1.75 inch long brass collimator and 0.5 mm lead pinhole to prevent saturation of the detector, even at low currents. The X-ray

source and detector were placed into a 61 x 46 x 42 cm³ lead-acrylic housing for safe irradiations.

E.1.2 Analysis

Measured spectra were imported into OriginLab's OriginPro energy analysis software. Regions of interest (ROI) were found using OriginPro's Quick Peak Gadget. The ROIs were then measured to determine the average energies and full-widths at half-maximum (FWHM) of the peaks. Wolfram Mathematica was used to visualize the data and compare the spectra.

E.1.3 Calibration

The X-ray detector was calibrated using Fe-55, Cs-137, Eu-152 and Am-241 sources and OriginLab's OriginPro energy analysis software. Each source was placed directly in front of the detector's collimator and the energy spectrum was measured for 180 s. The characteristic peaks of each source were marked as ROIs in the software and the proper energies were attributed to them using known spectra. The energy was then calibrated relative to the channel using a linear fit, allowing conversion from channel number to energy (Fig. E.1).

E.2 GAFChromic Radiochromic Film

GAFChromic EBT-3 radiochromic film was irradiated and scanned with an Epson Perfection V750 Pro high-resolution scanner to determine the diameter and uniformity of LuCIDD's X-Ray source's output beam. Prior to measurements with LuCIDD's X-Ray source, films were irradiated with a different, known X-Ray source for calibration measurements. After irradiation, the calibration films were cleaned and scanned. Measurements

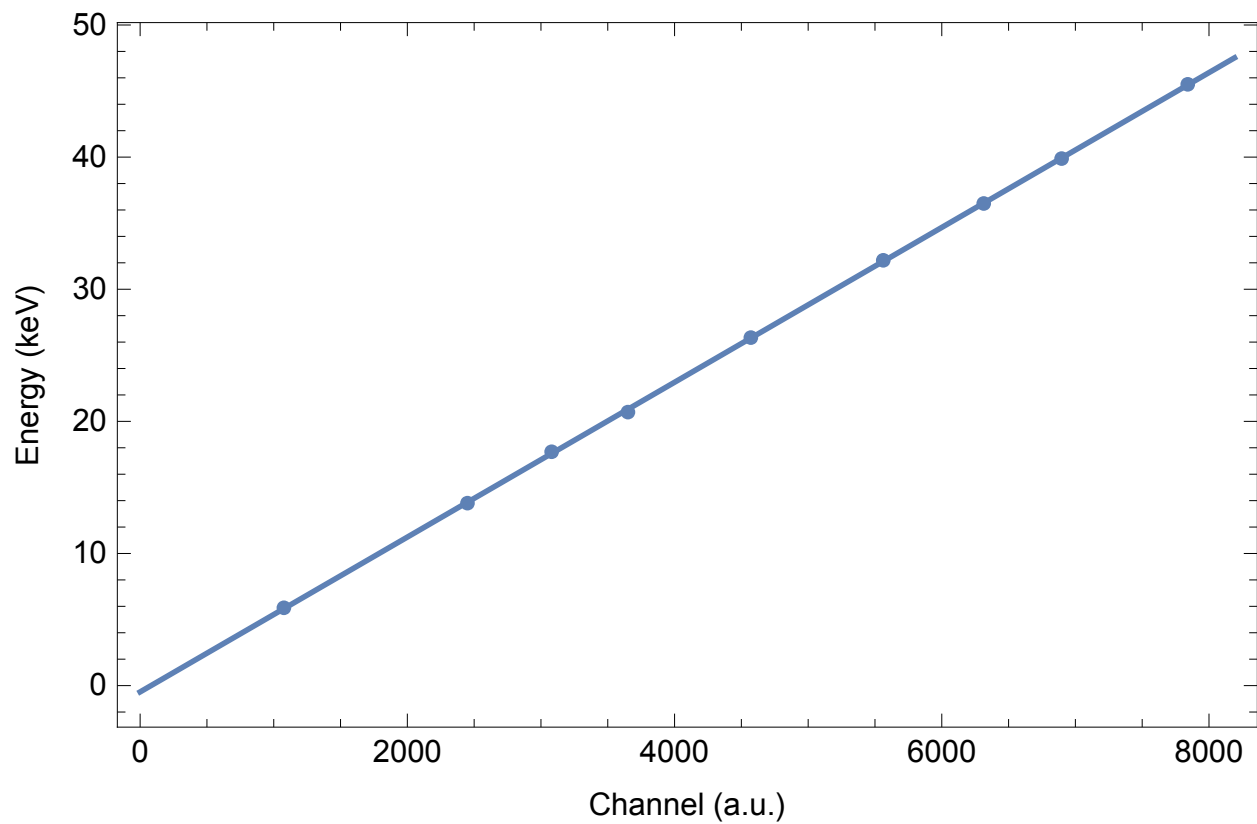


Figure E.1: The calibration curve for the X-ray detector was created by assigning known values to the peaks from multiple measure spectra, allowing energies to be assigned to each channel that the detector places its data.

were made on the scanned films using ImageJ and data was analyzed using Mathematica. This subsection describes the methods used to calibrate the GAFChromic EBT-3 radiochromic film.

E.2.1 Materials

EBT-3 film was obtained from GAFChromic for use with LuCIDD's X-Ray characterizations (Fig. E.2). EBT-3 film is comprised of a layer of active film about 28 μm thick that is inserted between two layers of clear, 125 μm polyester, and the film boasts an optimal dynamic dose range of 0.2 to 10 Gy (Howard, Herman, and Grams 2020). Prior to irradiation, films were cut and marked to ensure all films were scanned with the same orientation, due to known directional dependence when scanning film with document scanners. Once irradiated, the scanner and films were cleansed of fingerprints and other debris using denatured alcohol and a cotton swab. When not immediately in use, the films were stored in a light-proof container to prevent sunlight from darkening the sensitive films.

E.2.2 Instrumentation

The film was scanned into ImageJ using an Epson V750 Pro high-resolution scanner. The scanner was turned on at least an hour prior to use in order to warm up the electronics (Howard, Herman, and Grams 2020). To warm up the bulb, five "preview" scans were performed. The film was arranged down the center of the scanner to prevent lateral response artifacts (Lewis and Chan 2015). The film was scanned and imported into ImageJ, allowing measurement of the darkness of each pixel i.e., the mean grayscale value (MGV), which is inversely proportional to the dose absorbed by the film.

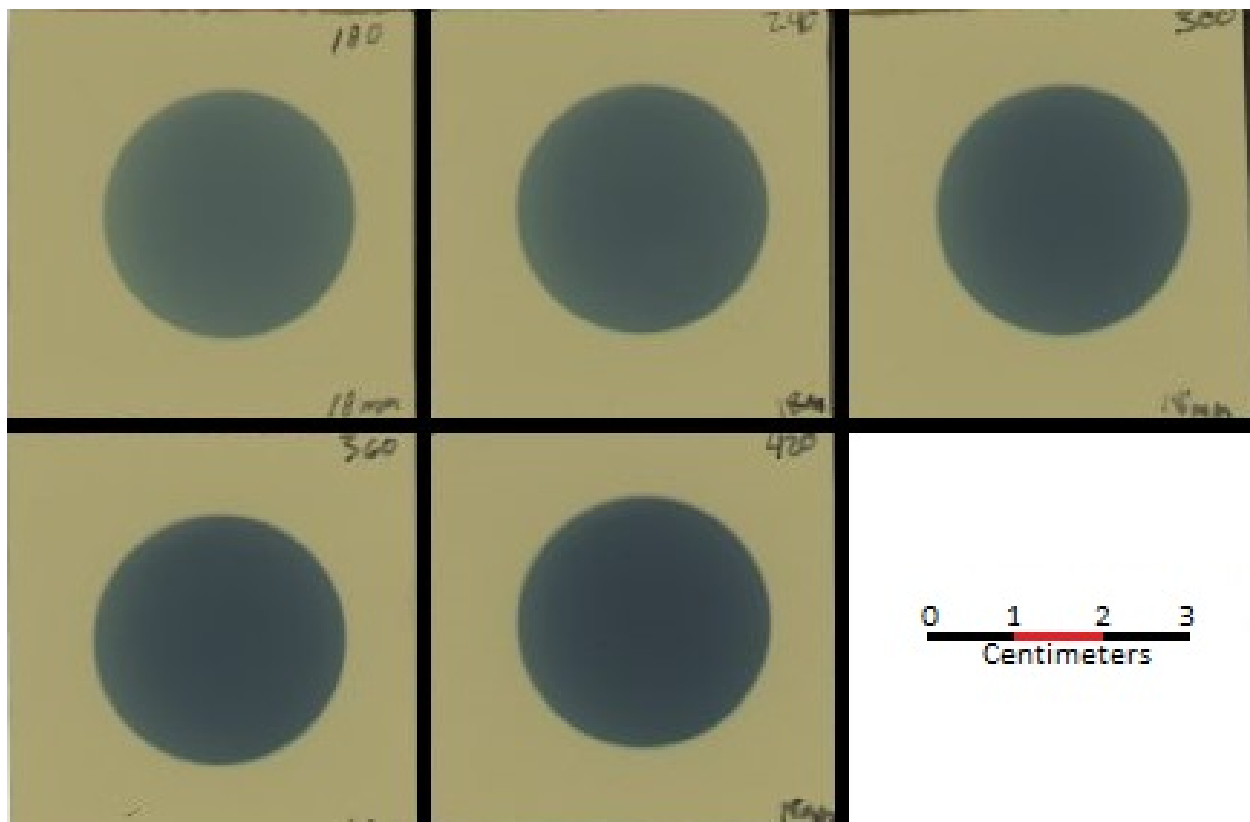


Figure E.2: A set of irradiated GAFChromic films. These films darken with increasing irradiation until reaching saturation and may be scanned and analyzed to determine the dose deposited to the film.

E.2.3 Analysis

Once measured, the MGVS of each film were imported into Mathematica and converted into dose using a calibration curve. The doses were visualized with three-dimensional dose-surface plots and two-dimensional cross-sections. Using these dose maps, the uniformity of the X-Ray's deposited dose were obtained at multiple beam radii.

E.2.4 Calibration

GAFChromic EBT-3 radiochromic film was calibrated using a 320 kV, 12.5 mA X-Ray source filtered with 2 mm of aluminum and at a source-to-sample distance of 50 cm. Multiple 5 cm strips were cut from sheets of film, marked for orientation, and irradiated with known doses ranging from 0 to 1500 cGy. The films were then left to develop for four days before being scanned with an Epson Perfection V750 Pro high-resolution scanner. These scans were imported into ImageJ where the MGVS of the combined channels, as well as those of the blue, green, and red channels were collected from 110 x 55 pixel ROIs centered on each strip (Howard, Herman, and Grams 2020). The MGVS were plotted against the known doses D and calibration curves of the form $MGV = a + b/(D - c)$ were fit to each channel (Fig. E.3).

E.3 Risø DA-20 TL/OSL Luminescence Reader

A Risø TL/OSL DA-20 luminescence reader was used to calibrate the effects of the X-ray source's dose rate relative to four sample types: quartz sediment, quartzite rock slices, sandstone rock slices, and Luxel dosimeter tape. Before X-ray characterizations could begin, the Risø beta source's dose rate was calibrated for each of these samples. Prior to this dissertation, the Luxel dosimeter tape was calibrated using a ^{137}Cs source.

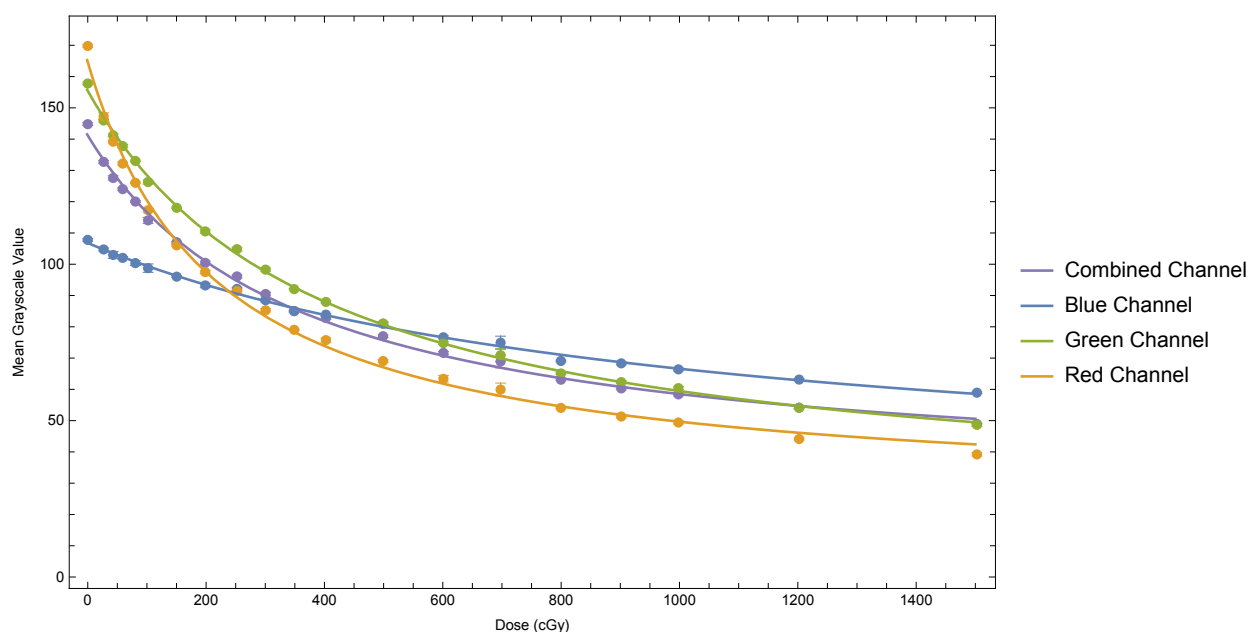


Figure E.3: GAFChromic film calibration curves for the combined, blue, green, and red channels. The red channel displays the most sensitive curve with the highest and lowest MGv values of any dataset. These curves are most accurate between 200 and 1000 cGy.

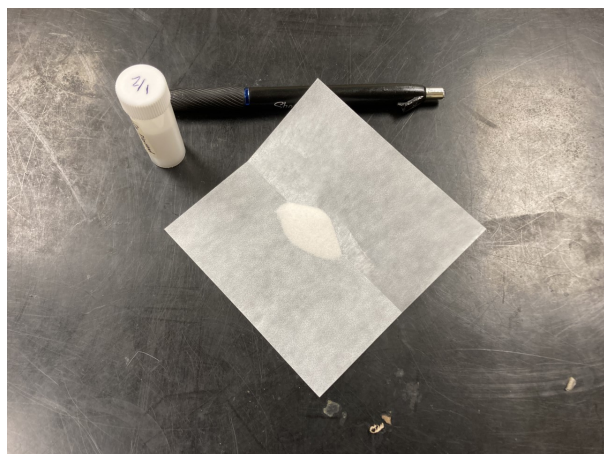
E.3.1 Materials

Commercial play sand was first sieved to grain sizes of 90 μm - 125 μm . The sieved sediment was then subjected to a density separation using heavy liquids. Density separation allows the isolation of quartz from any non-quartz minerals that could be a source of luminescence contamination, namely feldspars. The quartz fraction of the sediment was placed in an HCl bath to clean the sample of carbonate contaminants and etched with HF to dissolve any remaining feldspars. Finally, the quartz sediment was thermally reset by heating to 700 $^{\circ}\text{C}$ for one hour (Hansen et al. 2015). Aliquots of the quartz sediment were created by adhering grains to the surface of a stainless-steel cup using silicone spray (Fig. E.4a).

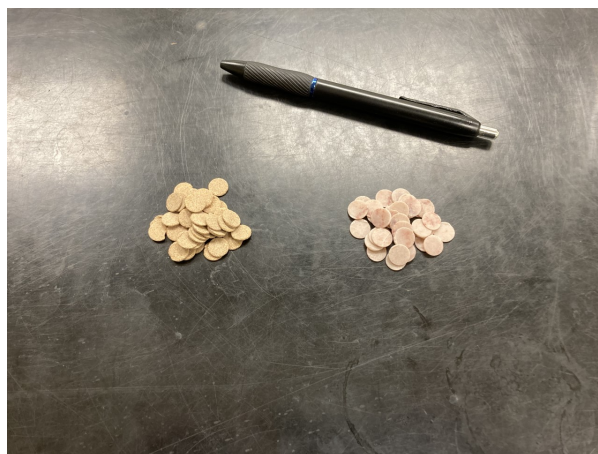
Two quartzite rocks and two sandstone rocks were obtained from Ward's Scientific for use with LuCIDD. Multiple 1 cm diameter cores were obtained from the rocks and were cut into approximately 1 mm thick slices along their cross sections (Fig. E.4b). The slices were also thermally reset by heating them to 700 $^{\circ}\text{C}$ for one hour. These slices fit well

into the Risø carousel and were each treated as an aliquot for the SAR procedure.

Luxel dosimeter tape was acquired from Landauer and processed into circular dosimeters using a 1/4 inch (6.35 mm) single-hole punch (Fig. E.4c). The Luxel dosimeters were optically reset by placing them under a halogen lamp filtered with a Kopp 3486 orange, long-pass filter for 24 hours (Yukihara et al. 2008). Each dosimeter was placed into individual stainless-steel cups to provide aliquots for measurement.



(a) Quartz sediment was processed for Risø and LuCIDD calibration measurements.



(b) Sandstone (left) and quartzite (right) rocks were cored and sliced to test the ability of the Risø and LuCIDD to date whole rock surfaces.



(c) Luxel dosimeter chips were created using a single-hole punch.

Figure E.4: The Risø reader's beta radiation source was calibrated for four samples.

E.3.2 Measurement and Analysis

The quartz sediment, quartzite, and sandstone were treated similarly during measurement by applying blue LED stimulation and a U-340 detection filter. The rock slices required a 10 mm aperture to be added to the detection filter to prevent PMT saturation due to the strength of their signals. Luxel was stimulated with green LEDs and a U340+L37 combination detection filter with 10 mm aperture (Fig. E.5).

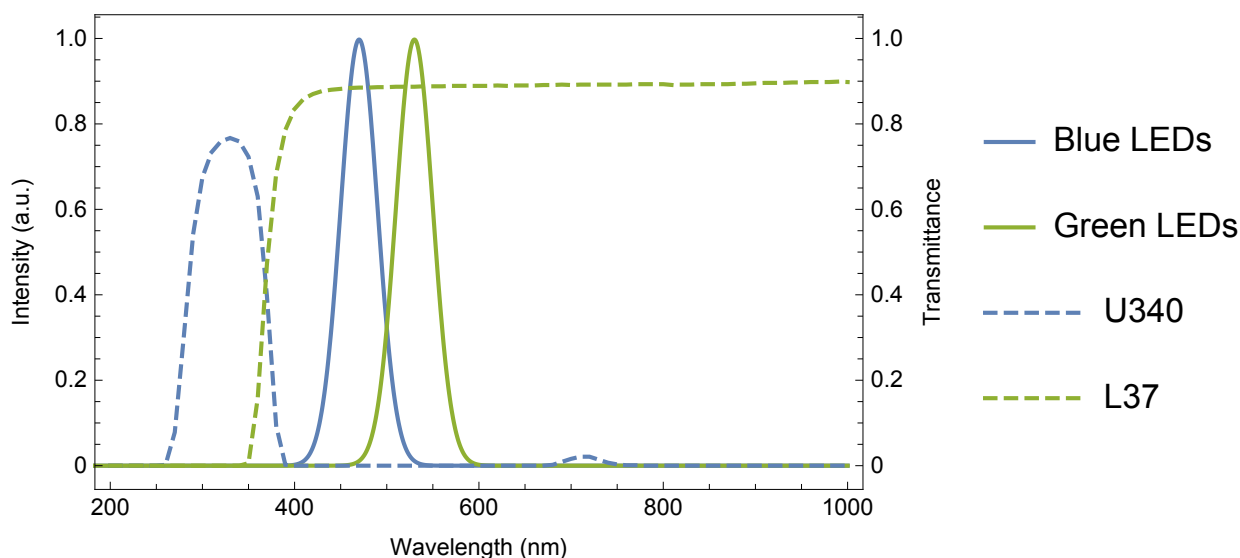


Figure E.5: Wavelengths for the blue and green LEDs and transmittances for their respective filters. When combined for Luxel measurements, the transmittance of the U340 + L37 filter becomes only the area underneath both of those curves.

Data analysis provided natural dose values either in seconds of beta (uncalibrated) or in Gy (calibrated) and was carried out using **R** and the “Luminescence” package (Kreutzer et al. 2012; Fuchs et al. 2015). The output from **R** was then analyzed and visualized using Wolfram Mathematica.

E.3.3 Suitability Testing of the Material

The Risø’s beta source was calibrated for dose rate relative to the quartz sediment, quartzite rock slices, and sandstone rock slices. Prior to this dissertation, the beta source

had been calibrated for Luxel dosimeter chips (Osunkwor 2022). To perform the required calibrations, plateau tests were performed, determining the optimal preheat temperature for each sample type (Fig. E.6). The preheat temperatures determined for the quartz sediment, quartzite rock slices, and sandstone rock slices were 190 °C, 210 °C, and 200 °C respectively. The doses recovered from the sandstone slices were consistently higher than the applied dose at all tested temperatures but fell within 10% of the applied dose throughout the plateau.

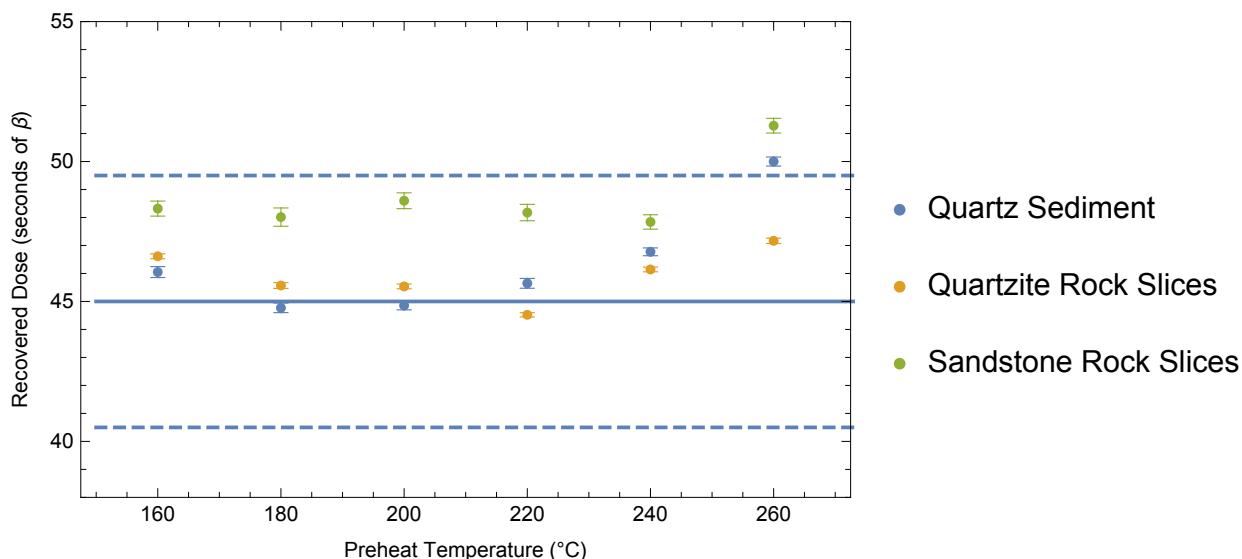


Figure E.6: Preheat plateau of quartzite rock slices. A “known” beta dose was given (in seconds) to multiple aliquots. The dose was recovered using the SAR procedure and the recovered value was compared to the preheat temperature. The solid line represents the applied known dose. The dashed lines represent the acceptable range ($\pm 10\%$) of the applied dose.

Once a preheat temperature had been determined for each sample, the accuracy of the SAR was tested by administering dose recovery tests (Fig. E.7). “Known” beta source doses from 0 to 210 seconds were given to each sample and the SAR protocol was performed using the previously determined preheat temperatures. Each sample passed the dose recovery test as the ratios of recovered doses to applied doses was found to be within 10% of unity for each sample. The dose recovery ratio for the quartz sediment, quartzite rock slices, and sandstone rock slices were 1.01, 0.95, and 1.06, respectively.

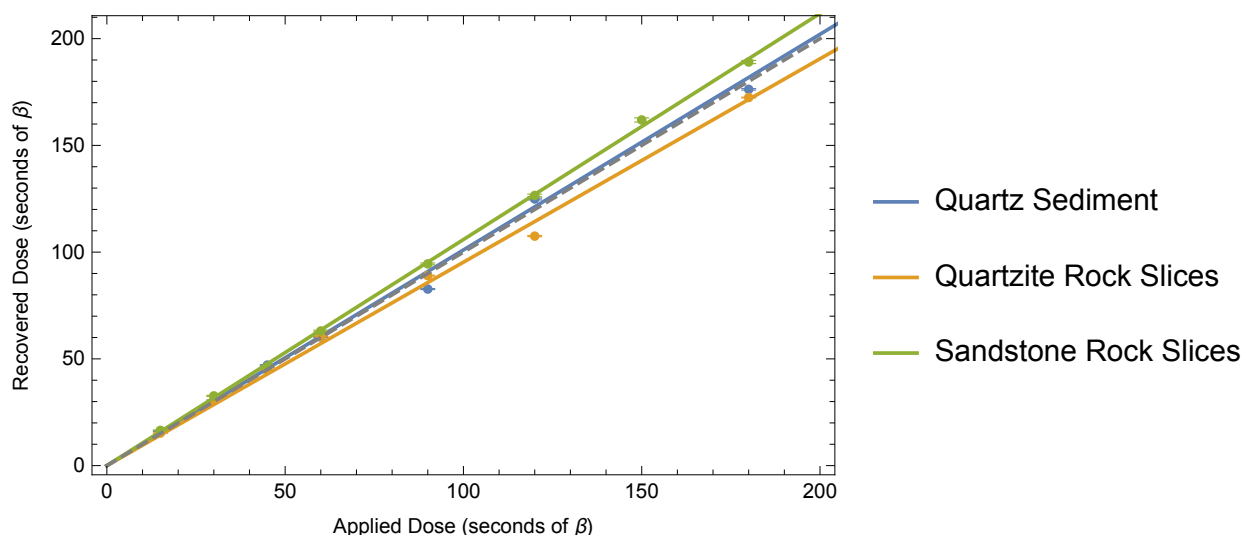


Figure E.7: Dose recovery test of quartzite rock slices. Multiple “known” doses were applied to different aliquots using the same preheat temperature. The doses were recovered using the SAR procedure and compared to the applied doses. Samples with ratios that are within 10% of unity are accepted.

E.3.4 Preparing Material for External Gamma Irradiation

After the SAR parameters were found and verified, the samples were packaged and sent for calibration irradiations. Quartz sediment was placed into two fused quartz vials and sealed with black, light-tight electrical tape. Packaging the rock slices required a second set of cores to be taken with a diameter of 1.65 cm, centered on previous cores. This resulted in thick cylindrical shells that were used as vials to hold the slices. Fine quartz powder was packed between the rock slices to prevent movement of slices during shipping and to maintain uniform density (Fig. E.8). Three quartzite vials and two sandstone vials were created this way. Once packaged, the samples were sent to an external facility for irradiations with a well-known Co-60 source. Due to the limited amount of aliquots for each sample, only a few different doses were applied: the two quartz sediment vials received doses of 5 and 7 Gy, respectively; the three quartzite rock vials received doses of 5, 7, and 10 Gy; the two sandstone rock vials received doses of 5 and 7 Gy. Two slices from the top and bottom of each vial were left unmeasured because we were unable to ensure surface effects wouldn’t alter their received dose relative to the interior slices.



(a) Samples prior to packaging.



(b) Packaged samples.

Figure E.8: Vials and light-proof, black electrical tape were used to package samples for calibration irradiations.

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