

MASTER'S THESIS, 60 ECTS

Construction of a hyperspectral Scheimpflug lidar for soot characterization in the aerosol phase

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Abstract

Greenhouse gases and light-absorbing particles in the atmosphere, including soot, play a key role in climate change. To understand the effect such small particles have on the climate, we first need to understand how they form and how they behave. Soot particles are produced from incomplete combustion processes of hydrocarbons, forming a mixture of graphitic and amorphous structures instead of carbon dioxide. Soot properties depend on combustion conditions, such as fuel type, temperature, and pressure. Many experiments have been conducted in a laboratory environment to understand the formation and properties of soot produced under different combustion conditions. Soot compositions can be inferred from their Raman spectra and the fluorescence emissions. Many spectroscopic techniques have been developed and applied widely in the lab scale, but not in naturally occurring conditions. For this reason, we aim to construct one for an atmospheric application based on the Scheimpflug principle and Raman scattering that is called Scheimpflug Raman Lidar (SRL). The Scheimpflug lidar technique has been used in many studies of insects, vegetation, and aquatic life. In the SRL, we use a high-power continuous-wave laser rather than a pulsed one. A simulation model was created prior to the theoretical model, which was optimized to resolve the Raman signal of soot. We successfully adapted the optimized construction and tested it on black soot (OP1 soot) and brown soot (OP6 soot) generated by a mini-CAST soot generator. To remove air contributions from these soot spectra, we combined SRL with polarization effects and Fast Fourier Transform (FFT) theory. By collecting data at various polarization angles, a FFT separated the unpolarized Raman signal of soot from the polarized air contributions. This split did not improve the Raman signal of soot, but it was able to identify all polarized contributions within the Raman spectrum, such as carbon dioxide, oxygen, nitrogen, and water vapor. This technique can thus be used to identify what gases are part of the combustion process.

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1 Introduction

1.1 Modern environmental studies

The United Nations has its 17 Global Sustainability Goals, which it aims to reach by 2030 [1]. Among these goals, some are related to by air pollutants, such as Goal 3: Good health and well-being; Goal 6: Clean water and sanitation; Goal 7: Affordable and clean energy; Goal 9: Industry, innovation and infrastructure; Goal 11: Sustainable cities and communities; Goal 12: Responsible consumption and production; Goal 13: Climate action; Goal 14: Life below water; Goal 15: Life on land. Given that over half the goals are relevant when studying air pollution, it is important to understand our sources of pollution and how they harm the health of our planet and the life on it, so that they can be remedied for a more sustainable future.

One type of pollutant that is particularly prominent in the atmosphere is soot particles. These are carbonaceous aerosols created from the incomplete combustion of hydrocarbons, typically fossil fuels, biomass, and waste burning. Soot particles are observed in the smoke from factory chimneys, car exhaust pipes, or any other type of combustion process. This means that industrialized and highly trafficked areas are particularly large sources of soot particles, which are polluting the air and saw a particularly large spike around the industrial revolution. The increasing presence of soot in the air is associated with deteriorating air quality. Soot particles are nanometer-sized, making them highly efficient at entering the bloodstream from the lungs upon inhalation. From there, they can cause inflammation and damage the respiratory system as well as cardiovascular functions [2].

On the environmental scale, the negative effects of soot particles become more apparent. Soot particles are good at absorbing sunlight, keeping many wavelengths trapped in the atmosphere that would otherwise escape. This raises the global average temperature [3]. Changes in temperature and reduced air quality harm many natural habitats and animal species, disrupting entire ecosystems [4]. More than that, soot particles in the air can be collected by precipitation and carried down to the surface [5]. This increases Arctic melting due to radiative forcing [6]. As Arctic sea ice melts, Arctic habitats change for many species, and sea levels rise [7, 8], harming coastal cities if the issue is not resolved. An increase in atmospheric soot also acidifies lakes, negatively affecting the habitat of fish and other aquatic species [9].

All these points lead to the natural conclusion that soot needs to be studied in detail, both in terms of how it forms and how it evolves, in order to do more accurate climate models. Since soot particles are very small and evolve continuously, a good practical way to study them is to use laser-based diagnostics in real time. Studying combustion processes that generate soot will provide insight into the formation of soot particles in the flame and how their molecular structure changes as they form under different conditions and mature in the aerosol phase. Acquiring this knowledge helps us take appropriate mea-

asures to reduce carbonaceous emissions that harm our planet and the life that resides on it.

1.2 Soot and its importance

Soot precursors, in its most basic description, consists of Polycyclic Aromatic Hydrocarbons (PAHs) [10]. They grow and form graphitic layers via intermolecular interactions, which then aggregate into large groups to form soot particles. Information about the molecular structure of soot can be obtained by measuring the Raman shift from inelastic scattering. Raman shift is the change in the frequency of an incident photon after an inelastic scattering, typically carrying information about the molecule’s vibrational structure. For instance, the peak intensity ratio of the Raman signal indicates the type of soot under investigation and its degree of maturity.

By studying different combinations of conditions in a lab, we build a library of knowledge of how soot particles can be identified. This helps studies conducted outside the laboratory, such as on house fires, forest fires, volcanic eruptions, and industrial processes, as well as for more accurate climate modeling to accurately assess climate impact. Having a foundation thus enables the identification of these soot-generation conditions, providing a clear picture of the full combustion process and the role of soot in the atmosphere.

A recent study shows that collecting soot on a substrate to study (called *ex-situ* measurements) does not yield the same results as measuring directly in the source (called *in-situ*, in-flight, or online measurements) [11]. Because of this, studies need to be conducted directly in the aerosol form for a more accurate analysis. This motivates the need to develop an experimental setup that measures the Raman spectra of soot particles directly in the smoke, potentially even at far distances. This would eliminate the need for direct access to the soot for sampling. By developing a remote sensing experimental setup, this issue will be addressed while also enabling the profiling of soot at different points in the source.

1.3 Scheimpflug lidar

Traditional lidar measures the round-trip time-of-flight (ToF) of pulsed lasers to determine distances. Since elastic scattering has the highest scattering cross section, it will give the strongest signal. In comparison, Raman signals are inherently weak, more than 1’000 times weaker than elastic scattering. Thus, if ToF ranging is to be used while collecting Raman signals from soot particles, the pulsed laser needs very high power pulses to get a detectable Raman signal. Unfortunately, this will cause thermal distortions or cause structural breakdown [12], meaning no coherent information can be extracted from a Raman spectrum.

Another lidar technique that does not require a high peak power laser is Scheimpflug lidar. Scheimpflug lidar instead uses optical geometry to determine distances through triangula-

tion rather than ToF, meaning it can use a Continuous-Wave (CW) laser for continuous signal sampling. A CW laser will not distort any soot particles, while still providing a strong enough Raman signal to extract information from. The basis of the Scheimpflug lidar technique is to have the transmitter at a slanted angle relative to the optical axis. This creates a tilted object plane, where each point on the plane is at a unique distance away from the receiver optics, allowing for unique mapping from source to detector. Thus, the distance is determined by the source's image position on the detector.

1.4 Roadmap of this thesis project

The Scheimpflug lidar technique has previously been used to study insects, vegetation, and aquatic life [13, 14, 15]. It has, however, not yet been used to study the Raman spectra of soot. The blueprints of the experimental construction were made by Oskar Öjstedt in a previous master's degree project [16], where the idea was confirmed to work in the simulated optical raytracing program ZEMAX. This thesis continues his work by focusing on the construction of the experimental setup and on the practical testing of the theory. Should a successful signal be recorded, the next step will be to attempt spatial and spectral calibration. If that is achieved, other tests can be performed to investigate how to remove Raman-active species in ambient air that overlap with the soot Raman signal, such as oxygen and carbon dioxide.

2 Theory

2.1 Light-matter interactions

Photons can be used to investigate soot particles, thus it is important to understand what type of interactions can be used to get information from. Two of these interactions that can give information about soot is fluorescence and Raman scattering.

2.1.1 Fluorescence

A photon interacting with a molecule can lead to a variety of outcomes, with probabilities governed by quantum mechanics that depend on the interaction conditions. One potential outcome is that a photon is fully absorbed by the molecule, leaving it in an excited state. The excited state of a molecule can be a combination of vibrational and electronic states, where electronic-state excitation requires more energy than vibrational-state excitation. An excited molecule will naturally want to relax to its ground state. This typically occurs by first relaxing between vibrational states via non-radiative transitions, such as thermalization. It will then relax between electronic states. If the transition is from a singlet state to the ground state, it is called fluorescence. If this transition is from a triplet state to the ground state, it is called phosphorescence. Figure 2.1 shows an energy-level diagram of these processes.

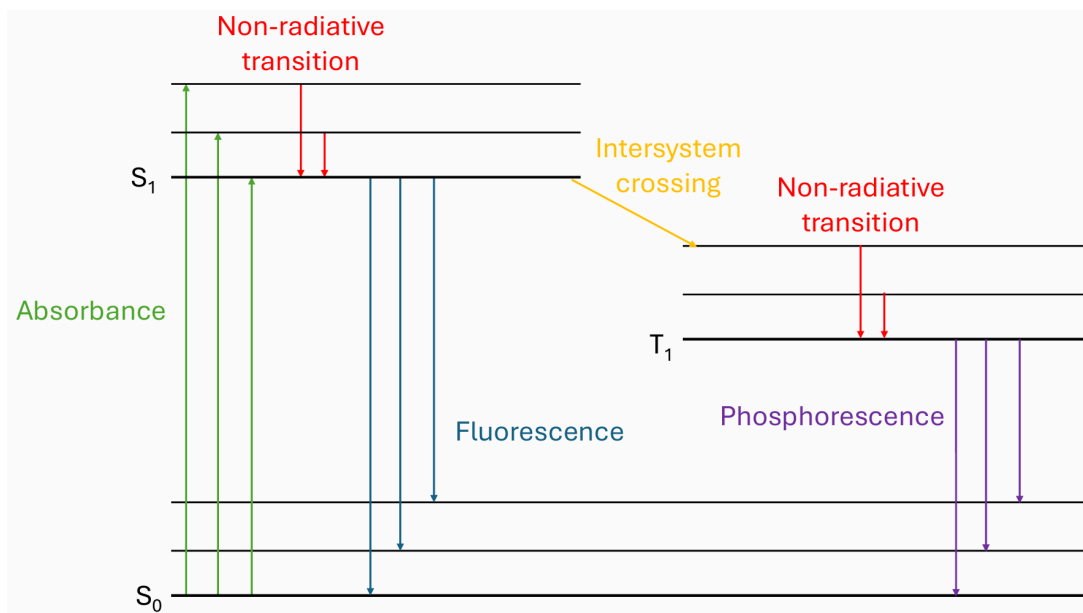


Figure 2.1: An energy level diagram showing different ways a molecule can relax from an excited singlet state S_1 to the ground state S_0 . First, it relaxes between vibrational states through non-radiative transitions. Then it either transitions directly from S_1 to S_0 via fluorescence, or it first intersystem-crosses to a triplet state T_1 and then relaxes to S_0 via phosphorescence.

2.1.2 Raman scattering

If a photon's energy is not precise for inducing an excitation in a molecule to a real excited state, it can instead be absorbed and induce a dipole moment in the molecule. This will excite the molecule to a virtual excited state. When relaxing to the electronic ground state, a photon can be emitted with either less, more, or the same energy as the incident photon by relaxing to a vibrational state that is either higher, lower, or the same as before the excitation. If it relaxes to the same vibrational state, it is called Rayleigh scattering. If it instead relaxes to a different vibrational state than the initial vibrational state, it is called Raman scattering. As shown in Figure 2.2, a relaxation to a higher vibrational state will emit a photon with less energy (longer wavelength), and this is called Stokes Raman scattering. If the vibrational state is lower, the photon will instead have more energy (shorter wavelength) and is called anti-Stokes Raman scattering. Raman scattering thus carries information about the vibrational structure of the molecule.

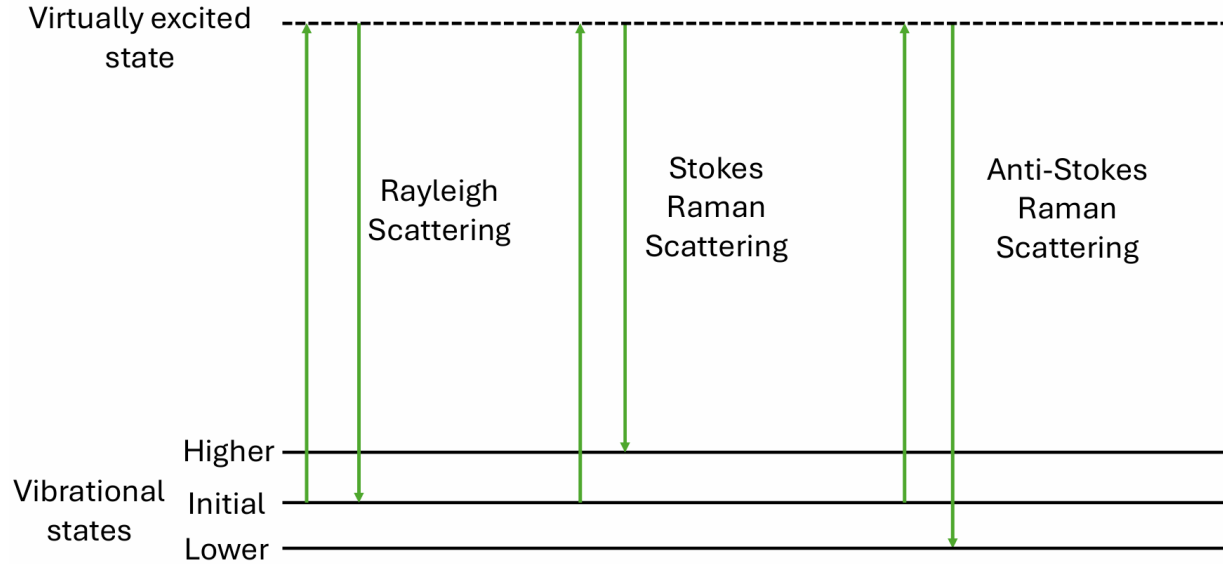


Figure 2.2: An energy level diagram showing three different scattering processes. A molecule in an initial vibrational state gets excited to a virtual state. Relaxation then occurs via anti-Stokes Raman scattering (more energy), Rayleigh scattering (same energy), or Stokes Raman scattering (less energy).

The energy of the scattered photon in the Stokes Raman scattering can be described in terms of frequencies of the scattered photon $\nu_{\text{scattered}}$, the incident laser photon, ν_{laser} , and the Raman shift of the transition ν_{Raman}

$$\nu_{\text{scattered}} = \nu_{\text{laser}} - \nu_{\text{Raman}} \quad . \quad (2.1)$$

Thus, when the laser and scattered photon frequencies are converted to wavelengths, the Raman shift ν_{Raman} of a laser wavelength, λ_{laser} is detected at wavelength $\lambda_{scattering}$.

$$\lambda_{scattered} = \left(\frac{1}{\lambda_{laser}} - \nu_{Raman} \right)^{-1}, \quad (2.2)$$

where ν_{Raman} is in units of wavenumbers. When constructing an experimental setup, Eq.(2.2) is used to determine which wavelengths to include in the detection interval.

2.2 Raman fingerprints of soot

The formation of soot particles is a continuous process that is very well, but not yet completely, understood [10]. When combustion occurs, carbon in the material bonds with oxygen in the air to form carbon dioxide. This is not always the case, as some carbons instead bind together to form polycyclic aromatic hydrocarbons (PAHs) [17, 18], meaning carbon rings that have bonded together into a 2-dimensional sheet [19]. In the early stages of formation, we call these precursors of soot [10]. Multiple of these sheets can group together to form layers, increasing the particle size [19]. The type of carbon rings that form soot particles determines the optical properties [20, 21]. A more organic compound gives larger fluorescence, and is called brown soot [10, 12]. As the particle matures, it contains fewer organic compounds as it becomes increasingly graphitic in structure. At this stage, it is called black soot and gives off less fluorescence [10, 12]. Figure ?? shows these two types of soot, with black soot in Figure ??a and brown soot in Figure ??b, together with TEM (Transmission Electron Microscopy) images of the soot particles on a molecular level. By looking at soot fluorescence relative to its Raman signal, it is possible to determine the maturity of the soot [21, 11].

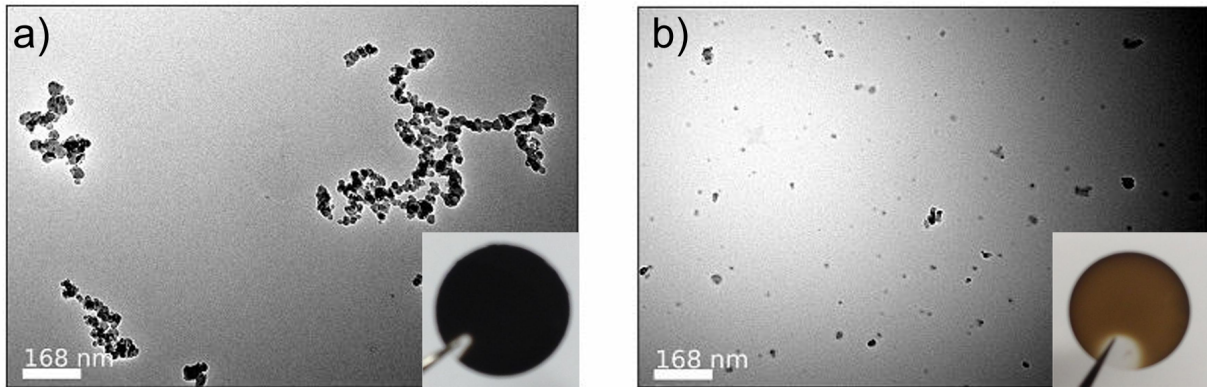


Figure 2.3: a) TEM image of black soot, with a macroscopic image in the bottom right corner. b) TEM image of brown soot, with a macroscopic image in the bottom right corner.

In a Raman spectrum of soot, there will be two dominant peaks called the D and G peaks [22]. The D peak is the A_{1g} symmetry vibrational mode of the carbon rings activated due to *disorder* caused by structural defects in the soot [23]. This is the so-called "breathing" vibrational mode, centered at a Raman shift of around 1360 cm^{-1} . The G peak, on the other hand, is caused by the *graphitization* of the soot, characterized by the E_{2g} -symmetric vibrational modes of the carbon rings [23]. This vibrational mode is called the "stretching" vibrational mode and is centered around a Raman shift of 1600 cm^{-1} . The intensity ratios between the D and G peaks carry information about the average graphitic crystalline size L_a [24]. The spectrum in Figure 2.4 from M.N. Ess et al. [25] shows these two characteristic peaks.

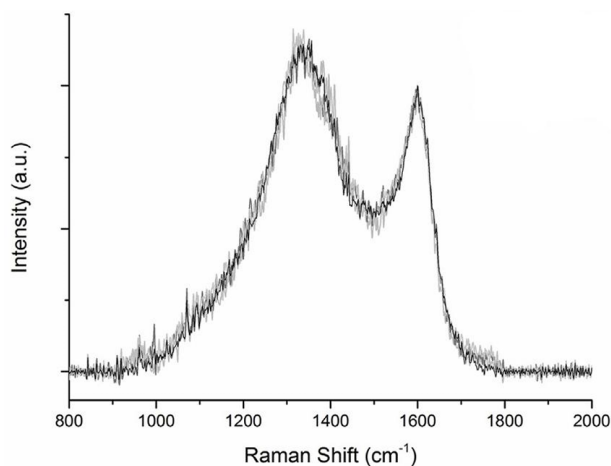


Figure 2.4: An example of a characteristic Raman spectrum of soot. The two dominant peaks visible are the D -peak at 1360 cm^{-1} and the G -peak at 1600 cm^{-1}

2.3 Air contributions

In the air, even in the absence of soot, there will still be molecules that have Raman signals close to and around the Raman signals of soot. Some of these are oxygen, nitrogen, and water vapor. Respectively, these will all be centered at 1555 cm^{-1} , 2331 cm^{-1} and 3654 cm^{-1} [11]. In the presence of soot, there will be carbon dioxide as well as a byproduct of the combustion, which will have two peaks at 1284 cm^{-1} and 1387 cm^{-1} [11]. These can be used to spectrally calibrate a detector, or act as indicators to the spectral detection interval the detector is set up to detect.

2.4 Experimental design

The complete experimental design is seen in Figure 2.5. Each part of the experimental design will be explained in the following sections from left to right.

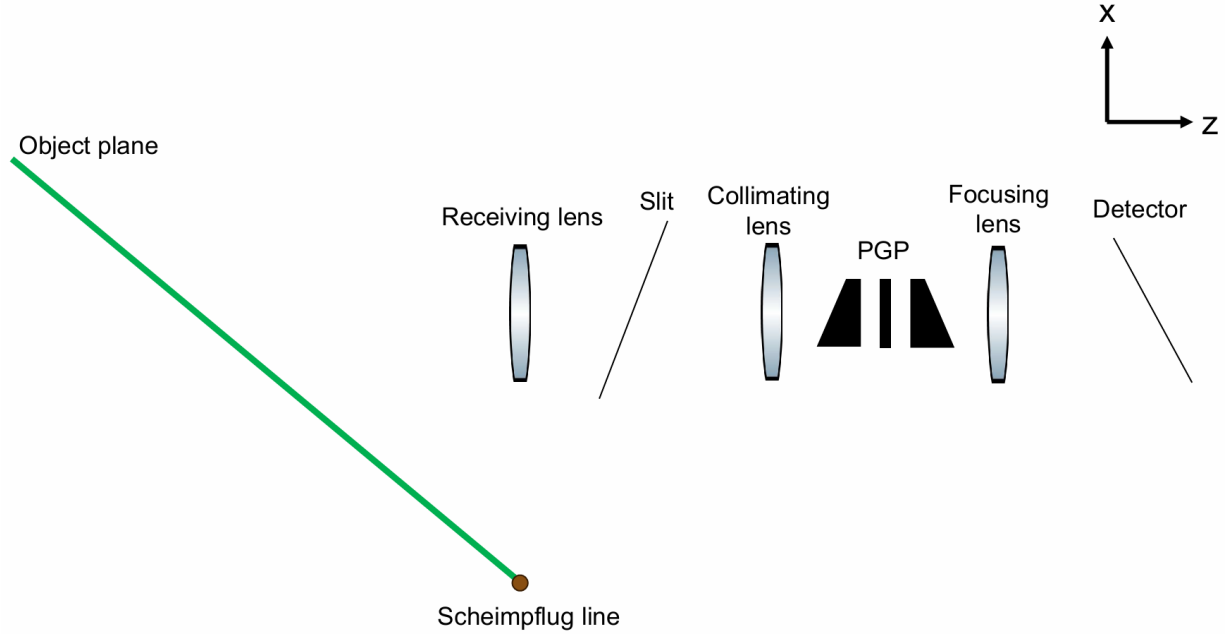


Figure 2.5: *Full schematic of the experimental design. An object plane will be imaged onto a slit through a receiving lens. The image will then get spectrally diffracted as it passes through the collimating lens, Prism-Grating-Prism (PGP) and focusing lens, ultimately being imaged onto a detector.*

2.4.1 Scheimpflug Lidar

Starting with the receiving lens and tilted image plane in Figure 2.5. The Scheimpflug lidar technique utilizes the geometry of the optical setup to determine the distance to a source located on the object plane. It combines two ideas to determine the image plane's location and orientation. The two ideas that are used together are the Scheimpflug principle [26] and the Hinge rule [27]. The Scheimpflug principle, demonstrated in Figure 2.6b, gives the position of the image plane. It states that a tilted object plane produces a tilted image plane, both of which intersect at the lens plane. The line of intersection is called the Scheimpflug line. The Hinge rule, demonstrated in Figure 2.6a, gives the orientation of the image plane by identifying two points of a *translated* image plane. It states that if a tilted image plane is translated so that it intersects the center of the imaging lens (first point), it will intersect the tilted object plane and the front focal plane on a common line (second point). This line is called the Hinge line.

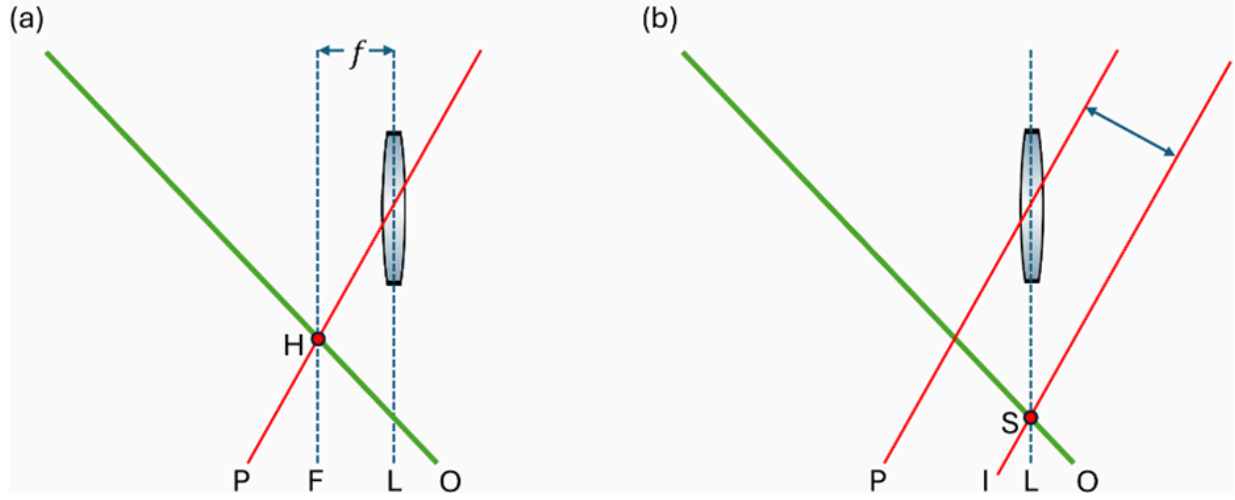


Figure 2.6: (a) *The hinge rule: The three planes - object plane O , the front focal plane F , and the translated image plane P that intersects the center of the imaging lens intersect at the Hinge line H .* (b) *The Scheimpflug principle: The three planes - image plane I , object plane O , and the lens plane L intersect at the Scheimpflug line S parallel to the previously translated image plane P .*

In the Scheimpflug lidar technique, a detector can be placed at the image plane of the laser beam. The transmitter should be placed on the Scheimpflug line, located a distance d from the optical axis on the lens plane. It is also from the Scheimpflug line that distances will be measured. The transmitter will be tilted at an angle φ relative to the optical axis to create the tilted object plane. This will make the image plane intersect the optical axis at a distance S from the imaging lens and tilted at an angle α relative to the lens plane. The transmitter parameters d and φ thus define the position S and tilt α of the image plane, which is illustrated in Figure 2.7. For an imaging lens with focal length f , these parameters are related to each other via

$$S = \frac{df}{d - f \tan(\varphi)} \quad (2.3)$$

$$\alpha = \tan^{-1} \left(\frac{f}{d - f \tan(\varphi)} \right) \quad , \quad (2.4)$$

with derivation provided in Appendix A.

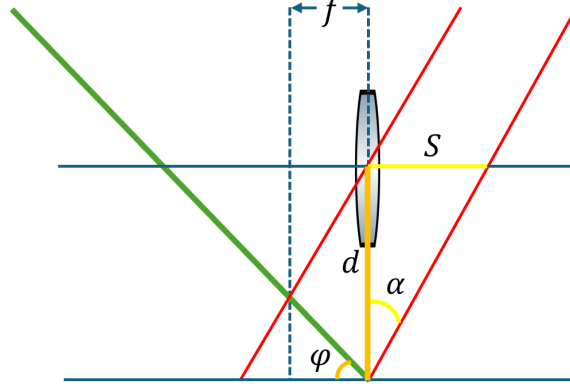


Figure 2.7: *Transmitter parameters (orange): The transmitter is placed a distance d along the lens plane from the optical axis and tilted an angle φ relative to the optical axis. Receiver parameters (yellow): The image plane will intersect the optical axis a distance S away from the imaging lens and be tilted at an angle α relative to the lens plane of the imaging lens.*

Typically, an experimental construction should be as simple as possible. A simple construction is possible if we let φ approach 0, and set $d = f$, then Eq.(2.3) and Eq.(2.4) simplifies the system to $S \approx f$, $\alpha \approx 45^\circ$. This approach is suited for constructing a setup with a fixed transmitter. If, instead, the receiver optics are fixed, one can find the position of the transmitter using

$$d = \frac{S}{\tan(\alpha)} \quad (2.5)$$

$$\varphi = \tan^{-1} \left(\frac{S - f}{f \tan(\alpha)} \right) \quad (2.6)$$

Eq.(2.5) demonstrates that d is independent of the focal length of the imaging lens. We defined d as the distance from the optical axis to the Scheimpflug line. Since the Scheimpflug line's position is independent of the imaging lens's focal length, it does not affect the line's location. If the receiver optics remain fixed, a change in the imaging lens's focal length will affect only the transmitter's tilt, enabling easier exploration of different range resolutions by simply switching the imaging lens.

Due to the range determination enabled by the Scheimpflug lidar method, it will be highly impractical to isolate any light source or scattering surface within the detector's Field Of View (FoV). Introducing a slit instead of a detector at the first image plane, as demonstrated in Figure 2.5 after the receiving lens, will allow the slit to act as a secondary source, cutting out interfering light outside the detectable range, blocking other sources within the detector FoV. Because the distance between a detected object and the receiver lens can be several meters and even kilometers, a collimating lens mere centimeters away from the slit can then easily collimate the light before it enters the Prism-Grating-Prism (PGP), which is very crucial for the PGP to operate as it is meant to and will be described in more detail later.

The imaging optics between the slit and the detector is the spectral separation optics, consisting of a PGP sandwiched between two lenses, a collimating lens and a focusing lens. Using two lenses can create a 4f-optical system if the distance between them equals the sum of their focal lengths. In such a system, an object located at the front focal plane of the first lens, such as the slit, will have its image at the back focal plane of the second lens, such as the detector, magnified by a factor M_{4f} defined by

$$M_{4f} = -\frac{f_{foc}}{f_{col}} \quad , \quad (2.7)$$

where f_{col} is the focal length of the collimating lens and f_{foc} is the focal length of the focusing lens. Approximating the full slit to be at the front focal plane of the collimating lens, the length of the slit, l_{slit} , can be determined by the length of the detector, l_{det} , and the absolute magnification of the 4f-optical system

$$l_{slit} \approx \frac{l_{det}}{|M_{4f}|} \quad . \quad (2.8)$$

The absolute value of the magnification is used because the detector and slit sizes are non-negative. Similarly, the angle of the detector, α_{det} , is related to the tilt of the slit α_{slit} via

$$\alpha_{det} \approx \alpha_{slit} M_{4f} \quad . \quad (2.9)$$

2.4.2 Prism-Grating-Prism

A Prism-Grating-Prism (PGP) is a tool for spectral separation of light. The benefit of including a prism before the grating is that it increases its dispersion efficiency while maintaining the same optical axis as the prior optics. First, the light needs to be collimated before it enters the first wedge prism of the PGP. Once the light is collimated, it will pass through an iris to shape the beam and remove all surrounding light. The prism will bend the collimated light off the optical axis by the Bragg angle θ_B of the grating for some wavelength λ_B . The wedge angle β_1 of the prism is what decides the angle by which the incident light gets bent, meaning it will have to be carefully chosen¹. Since the light will be incident at the Bragg angle θ_B of the grating for some wavelength λ_B , it will mean that the diffraction at this wavelength from each groove separated by a distance d will experience complete constructive interference at different orders m since it fulfills the Bragg condition:

$$m\lambda_B = 2d \sin(\theta_B) \quad . \quad (2.10)$$

All wavelengths around λ_B will approximately fulfill the Bragg condition for almost complete constructive interference after diffraction. The second wedge prism, after the grating,

¹The procedure to find this angle can be found in Mauri Aikio's doctoral dissertation [28].

will realign the $m = -1$ order with the prior optical axis. The wedge angle β_2 of the second prism is chosen similarly to β_1 .

A lens after the second prism focuses the diffracted light onto a detector. This lens determines the spectral range that the detector can detect. The relation between the height of the detector in the spectral dimension H_λ , the detector assembly coefficient a , the grating density ν , the focal length of the focusing lens f_{foc} , the central wavelength λ_C of the detection interval, and the longest wavelength λ_L of the detection interval is

$$\nu(f_{foc}) = \frac{1}{\sqrt{\left(\frac{\lambda_L}{Q}\right)^2 - \frac{2W\lambda_L\lambda_C}{Q^2} + \left(\frac{W\lambda_C}{Q}\right)^2 + \lambda_C^2}}, \quad (2.11)$$

where the parameters Q and W are defined by

$$Q = \frac{aH_\lambda}{2f_{foc}\sqrt{1 + \frac{aH_\lambda}{2f_{foc}}}} \quad (2.12)$$

$$W = \frac{1}{\sqrt{1 + \frac{aH_\lambda}{2f_{foc}}}}, \quad (2.13)$$

which are derived in Mauri Aikio's doctoral dissertation [28]. By choosing a detector, grating density, and wavelength range, the focal length of the focusing lens can be determined to achieve the desired spectral range. Figure 2.8 provides a full illustration of how it all comes together as light travels along the optical axis (the z -direction).

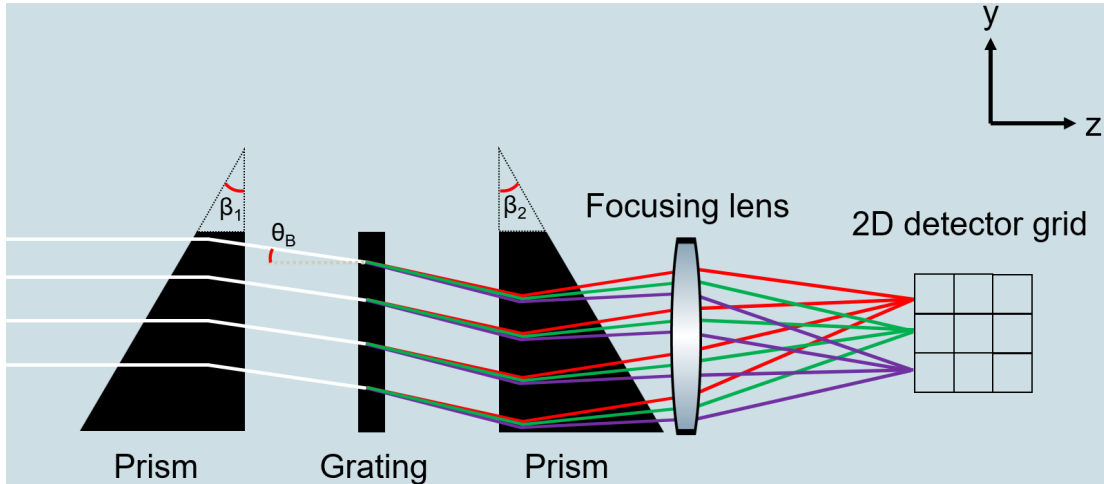


Figure 2.8: White collimated light enters the PGP and gets diffracted where different wavelengths get focused onto a detector grid at different heights. The wedge angle β_1 is picked so that θ_B is the Bragg angle for some wavelength. The wedge angle β_2 is chosen to re-align the light with the optical axis.

Filters can be included to avoid interference between too many wavelengths. When looking at Stokes Raman scattering, a long-pass filter centered around the laser wavelength removes the anti-Stokes components, and a notch filter centered at the laser wavelength removes the elastic scattering. The remaining signal will thus only be the Stokes Raman Scattering.

When combined with the Scheimpflug principle, which spatially separates the light horizontally along a line, each point along the line will get spectrally diffracted vertically, creating a 2D image. On a 2D detector grid, the image observed by the detector is shown in Figure 2.9, where we are looking down the z-axis (the optical axis).

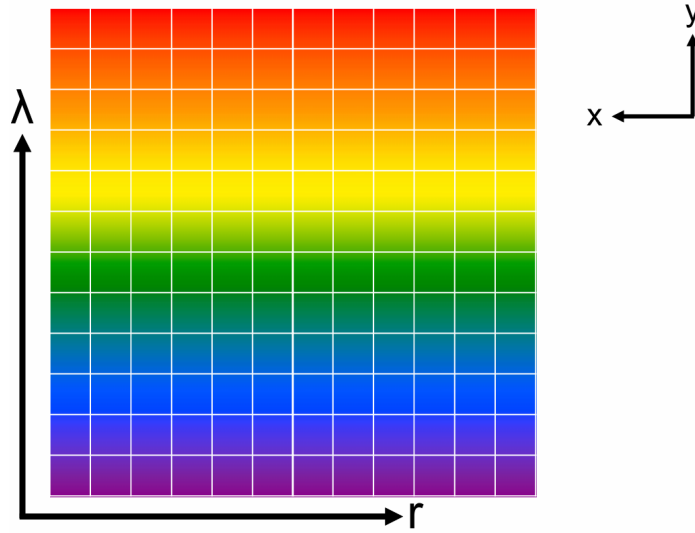


Figure 2.9: Image seen on the detector after light from the object plane has been spatially separated horizontally along the x -axis, and spectrally separated vertically along the y -axis.

2.4.3 Ranging

Since the Scheimpflug principle is used twice in the experiment to find two distinct image planes, a formula to determine the ranges based on where a signal is registered on the detector must account for this. The object plane from a transmitter tilted at an angle φ relative to the optical axis will be imaged by a receiver lens with focal length f_{rec} on the slit, which is tilted at an angle α_{slit} relative to the lens plane shown in Figure 2.7. The image of the slit will be formed on the detector via the 4f-optical system with magnification M_{4f} . One point on the object plane will be mapped to one pixel column p_r on the detector, and each pixel has a length of l_{pix} . The end $p_r = 0$ represents the farthest detectable distance, and the end $p_r = p_{r,max}$ represents the closest detectable distance. The formula to relate the pixel column p_r to a measured distance r is

$$r(p_r) \approx \frac{f_{rec}}{\cos(\varphi)} \left(1 + \frac{|M_{4f}| f_{rec}}{|M_{4f}| (b - f_{rec}) + p_r l_{pix} \sin(\alpha_{slit})} \right), \quad (2.14)$$

with derivation provided in Appendix B. The parameter b is a calibration parameter. The derivation of Eq.(2.14) leaves room for the horizontal center of the detector to not be fully aligned with the optical axis. The definition of b is the horizontal distance from the receiver lens's lens plane to the position on the slit being mapped to $p_r = 0$ on the detector, which is visually demonstrated in Figure 2.10 for clarity. If the detector is translated along the 2nd image plane, it will change the detector's FoV and the value of b . Hence, the only parameter that needs to be calibrated is dependent on the detector's position.

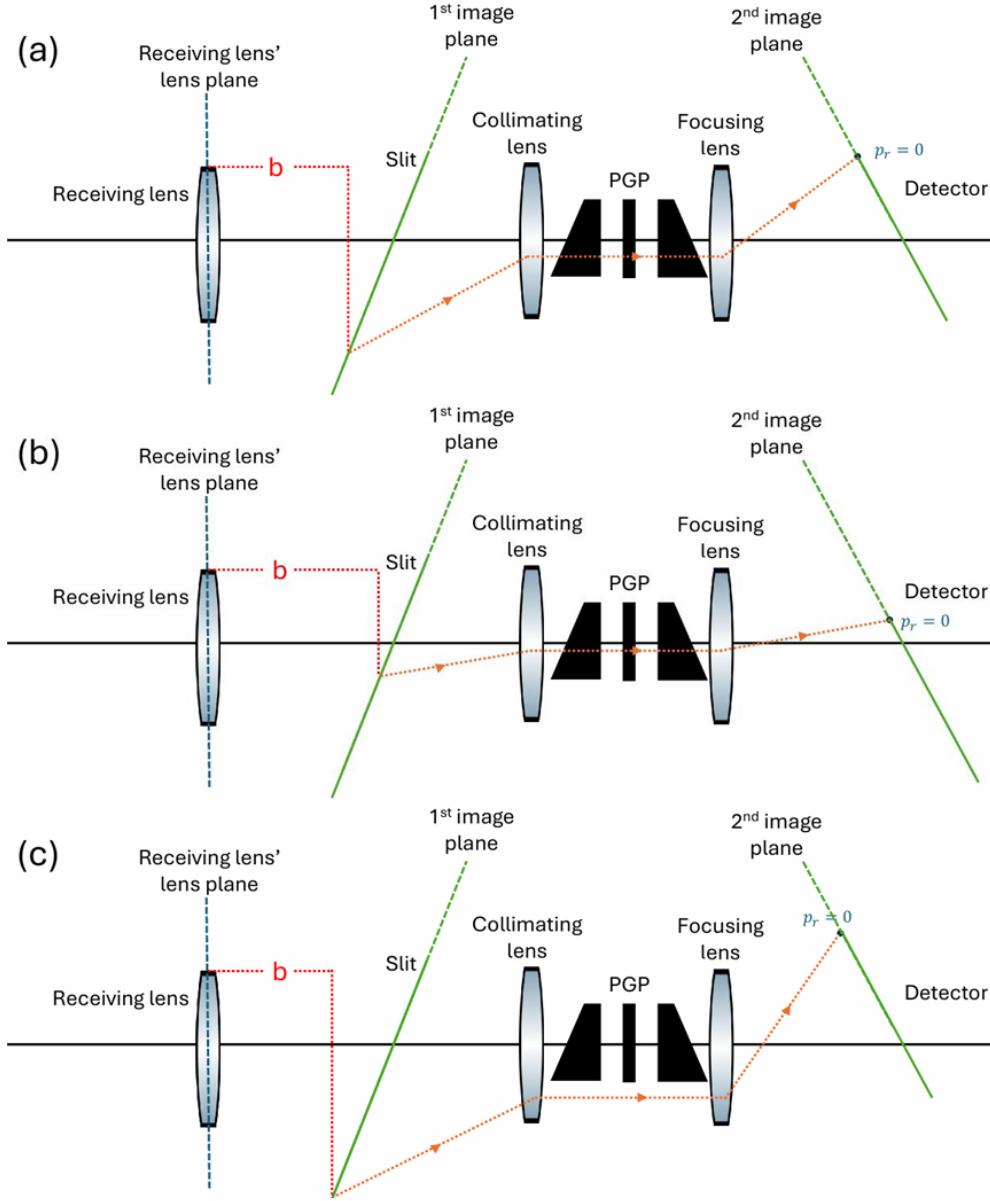


Figure 2.10: A top-view schematic of how b in Eq.(2.14) is defined, and how it is dependent on the position of the detector along the image plane. (a) The detector's center is aligned with the optical axis. (b) and (c) The detector's center is not aligned with the optical axis.

One very important consideration to account for is that every range between $r(p_r - 1)$ and $r(p_r)$ will be mapped to the pixel column p_r , which can be understood by Figure 2.11. The consequence of this is that there will always be an unavoidable uncertainty in range measurements. Due to the nature of Eq.(2.14), this uncertainty will decrease as p_r increases, meaning a small uncertainty at closer distances and a larger uncertainty at longer distances. For measurement purposes, the range for $r(p_r - 1)$ and $r(p_r)$ should both be determined to give a range interval, where $r(p_r)$ is the closest possible distance and $r(p_r - 1)$ is the farthest possible distance.

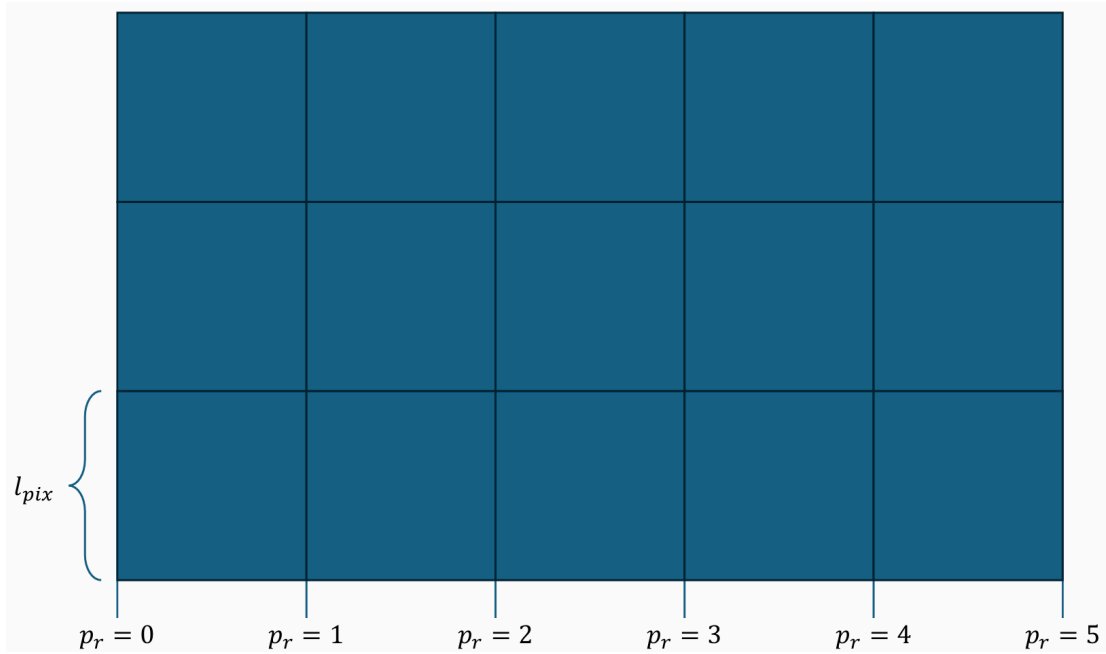


Figure 2.11: A portion of a detector 2D pixel surface. The pixel column p_r is defined at the right edge of a column, and the pixel size is l_{pix} . All distances between $r(p_r - 1)$ and $r(p_r)$ will be mapped to pixel column p_r .

2.4.4 Ranging calibration

Since b needs calibration, a single reference point needs to be measured. Due to the increasing range uncertainty interval $r(p_r - 1) - r(p_r)$ coinciding with a pixel column at longer ranges, the calibration point should be as close as possible within the detection range, where the uncertainty interval is small. Some reflective surface should be placed at a known, measured distance r_{ref} from the Scheimpflug line along the beam path, which, when illuminated by the transmitter, will be mapped to the detector pixel column $p_{r,ref}$. It is then straightforward to solve for b in Eq.(2.14) and use it to determine the range interval for any pixel column.

2.4.5 Spectral calibration

The PGP will spectrally separate the light vertically, along pixel rows p_λ , whereas the spatial separation from the Scheimpflug technique is horizontal, across pixel columns p_r . This creates a 2D image with distance along the x-axis, wavelength along the y-axis, and intensity on the z-axis. Each pixel must be uniquely calibrated by identifying known peaks in a recorded image. The wavelength dependence on a pixel $\lambda(p_\lambda, p_r)$ is a polynomial of p_λ and p_r . The calibration function used in this project is quadratic with respect to pixel row, p_λ , and linear with respect to pixel column, p_r .

$$\lambda(p_r, p_\lambda) = a_0 + a_1 p_r + a_2 p_\lambda + a_3 p_r p_\lambda + a_4 p_\lambda^2 + a_5 p_r p_\lambda^2 \quad . \quad (2.15)$$

2.5 Separating polarized components from unpolarized components

2.5.1 Discretised Fourier Transform

The Fourier Transform of a periodic function $f(x)$, which can be in a superposition of multiple periodic functions of unknown periodicities, amplitudes and phases, will transform into $\tilde{f}(k)$. In the new domain, the periodic functions will be separated by their frequencies, and have transformed amplitudes relative to their oscillating amplitudes, and can thus be identified. However, if $f(x)$ doesn't have a mathematical expression, then one cannot perform the traditional Fourier Transform. Instead, one can collect data points for $f(x)$ and then do what is called a Discretised Fourier Transform (DFT), also known as a Fast Fourier Transform (FFT), defined as

$$\tilde{f}(k) = \sum_{n=0}^{N-1} f(x_n) e^{-i \frac{2\pi}{N} kn} \quad , \quad (2.16)$$

where N is the number of data points recorded for $f(x)$.

Let us say a spectrum $f(x)$ itself is not oscillating, but has some function values that oscillate as some parameter θ changes. Then data can be collected for $f(x, \theta_n)$ at different θ_n . Once a set of $f(x, \theta_n)$ has been recorded, one can perform the FFT using all $f(x, \theta_n)$ to get $\tilde{f}(x, k)$. The FFT in such a case is defined similarly to Eq.(2.16) as

$$\tilde{f}(x, k) = \sum_{n=0}^{N-1} f(x, \theta_n) e^{-i \frac{2\pi}{N} kn} \quad . \quad (2.17)$$

Each $f(x, \theta_n)$ need to be sampled with stepsizes $\Delta\theta$ ranging over a full period, meaning that N is defined as

$$N = \frac{360^\circ}{\Delta\theta} \quad , \quad (2.18)$$

hence n will go from 0 up to $N - 1$ and θ goes from 0 to $(N - 1) \Delta\theta$.

The magnitude of the FFT can be normalized by N ,

$$\tilde{F}(x, k) = \frac{|\tilde{f}(x, k)|}{N} \quad , \quad (2.19)$$

for easy extraction of the desired values.

2.5.2 Simplest application of the FFT

Assuming a signal only has dependence on one oscillating function, it can be split into two simple components: the average term and an oscillating term that oscillates around the average with some amplitude. Such a function can be described as follows:

$$f(x, \theta_n) = f_{av}(x) + f_{AC}(x, \theta_n) \quad , \quad (2.20)$$

which is more clearly illustrated in Figure 2.12a at some arbitrary x that has a visible oscillating intensity as a function of θ with amplitude $A(x)$.

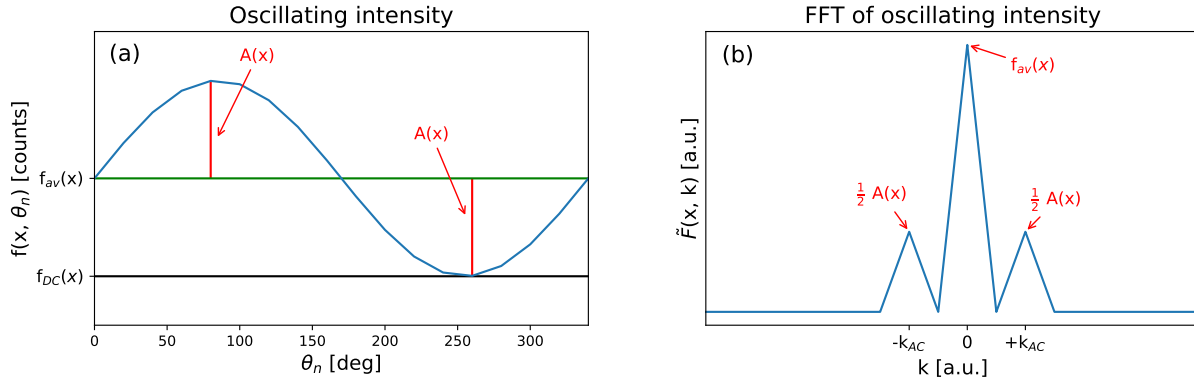


Figure 2.12: (a) A full period of an arbitrary oscillating function $f(x, \theta_n)$ that oscillates as a function of θ_n around $f_{av}(x)$ with amplitude $A(x)$ and minimum value $f_{DC}(x)$. (b) The normalized FFT of $f(x, \theta_n)$, $\tilde{F}(x, k)$, with $\frac{1}{2}A(x)$, and $f_{av}(x)$ marked as the spike amplitudes.

When Eq.(2.20) is substituted into Eq.(2.17) and normalized using Eq.(2.19), one will find that the $k = 0$ term will look like this:

$$\tilde{F}(x, 0) = f_{av}(x) \quad . \quad (2.21)$$

The oscillating values can be removed from $f(x, \theta_n)$ and only leave what is constant across all $f(x, \theta_n)$. We will call this the DC term $f_{DC}(x)$. We can easily derive an expression for $f_{DC}(x)$ by looking at Figure 2.12a where we see that

$$f_{DC}(x) = f_{av}(x) - A(x) \quad . \quad (2.22)$$

The amplitude $A(x)$ of $f_{AC}(x, \theta_n)$ can be determined from $\tilde{F}(x, k)$. The amplitude will be split into two symmetrical peaks in $\tilde{F}(x, k)$, which are marked in Figure 2.12b as $\frac{1}{2}A(x)$. These two peaks are located at $k = -k_{AC}$ and $k = +k_{AC}$, respectively. Thus, $f_{DC}(x)$ at each value of x , can be determined from the FFT of $f(x, \theta_n)$ by

$$\begin{aligned} f_{DC}(x) &= f_{av}(x) - A(x) \\ &= \tilde{F}(x, 0) - 2 \cdot \tilde{F}(x, k_{AC}) \quad , \end{aligned} \quad (2.23)$$

which is uniquely calculated for each x . The resulting $f_{DC}(x)$ spectrum will have removed any terms that oscillate in intensity because of a change of some periodic parameter θ .

The oscillating terms, $2 \cdot \tilde{F}(x, k_{AC})$, can then be plotted separate from $f_{DC}(x)$ to show all components removed. Such a plot can be used to identify previously hidden information in $f(x)$ that was not previously identifiable because it was masked behind a much stronger DC signal.

3 Method

3.1 Constructing the transmitter

Oskar Öjstedt provided blueprints for the experimental setup in his master's degree project [16]. The transmitter was constructed using an elliptically polarized 532nm CW laser with a maximum power output of 2W. Attached to the laser is a video objective with a tunable focus. The video objective consisted of a negative lens, a positive lens, and an iris. The distance between the two lenses enables change in focal length, and the iris shapes the beam. The video objective was necessary because the laser's Gaussian beam diverged too quickly without it. It is crucial that as many interactions as possible occur within a small probe volume. So the beam waist radius should be kept as small as possible around the sample. The tunable focus changes the focal point and, consequently, the beam divergence, so that it is at a minimum at close ranges. Not only does this increase the number of interactions, but it also ensures that the interactions pass through the slit entrance and can be detected. Since the beam is imaged onto a slit, any interaction that gets imaged above or below the slit entrance will be lost, decreasing the photon flux through the slit. This is why the beam size cannot be too large, hence the implementation of the video objective to slim it down.

The transmitter was mounted on a rotating platform so that the slant angle could be set to $\varphi = 5.5^\circ$. In turn, the rotating platform was mounted on a height-adjustable platform so that the transmitter's height could be aligned with the receiver's optical axis. The full transmitter construction is shown in Figure 3.1.

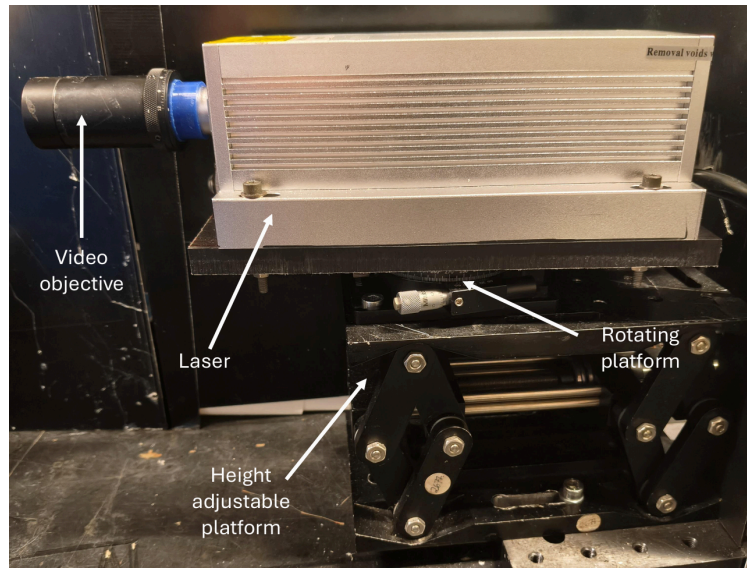


Figure 3.1: *The constructed transmitter, consisting of a 532nm CW laser with an attached video objective. The laser is mounted on a rotating platform and a height-adjustable platform.*

An attempt was made to add a second rotator for vertical rotation of the transmitter, but the laser was too heavy for such a construction, and no alternative method compatible with an adjustable-height platform was found. The purpose of such a vertical rotator is to make the laser beam horizontal with respect to the platform on which it is mounted. This is because the laser, by itself, did not produce a horizontal beam but a beam tilted slightly downwards. This issue was instead fixed by the video objective, as it turned out that it could be used to direct the laser beam without moving the laser. The downside to using the video objective to direct the laser beam is that it is sensitive and not quantifiable. Guide marks in the form of irises needed to be placed in the beam path where the beam would be horizontal, and the video objective was then adjusted so that the beam passed through all the irises simultaneously.

3.2 Constructing the receiver optics

As part of the receiver optics, the following components were mounted on a dovetail optical rail to achieve a common optical axis:

- A $\varnothing 2''$, $f_{rec} = 200$ mm receiver lens,
- a 50×0.25 mm² slit,
- a $\varnothing 2''$, $f_{col} = 150$ mm collimating lens,
- a 18.15° wedge angle prism,
- a 600 grooves/mm grating
- a 18.15° wedge angle prism,
- a notch filter,
- a long pass filter,
- a $\varnothing = 1''$, $f_{foc} = 80$ mm focusing lens.

The receiver and collimating lenses were mounted in the same type of optical holders on a dovetail optical rail, ensuring they maintain the same optical axis. The PGP, filters, and focusing lens were placed inside of a $\varnothing 1''$ lens tube, which was connected to the back end of the collimating lens mount, again so that they all share the same optical axis. The dovetail optical rail was also, similarly to the transmitter, fastened to a large height-adjusting platform as seen in Figure 3.2.

After the optical rail was a 2D Andor CCD camera with a 512×2048 -pixel grid. Each pixel has a width of $l_{pix} = 13.5 \mu m$. In the spatial (horizontal) dimension, the width of the detector pixel grid is $l_{det} = 2048 \cdot l_{pix} = 27.6$ mm. The magnification of the 4f-optical system was determined by Eq.(2.7) to be $M_{4f} = -0.533$, hence the width of the slit

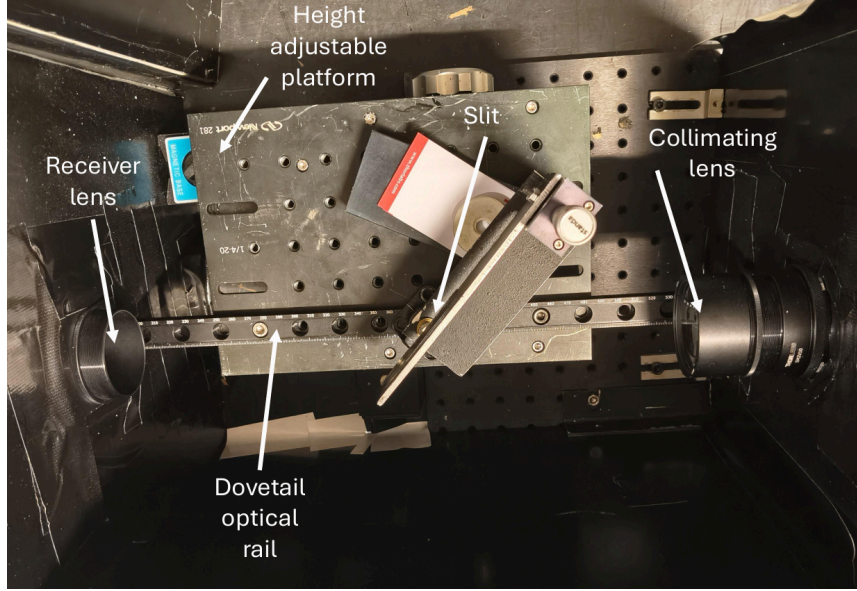


Figure 3.2: The dovetail rail with receiver lens, slit, and collimating lens mounted on it.

was determined by Eq.(2.8), which became $l_{slit} \approx 50$ mm. No such slit was available for ordering, so the slit had to be constructed manually. It was made with two box-cutter razor blades superglued to a piece of metal with an entrance width of 50 mm. The space between the razors was separated by a 0.25 mm thick metal sheet. The slit entrance thus has dimensions of 50×0.25 mm². Ideally, the slit would have a height of 0.1 mm, but no such thin metal sheet was available. The metal plate with the razor blades glued to it was mounted on a miniature height adjuster. The miniature height adjuster allows for micrometer-precise control of the height of the slit, which was tuned to make sure the slit entrance was aligned with the optical axis. The mini height adjuster was placed on a small platform, which in turn was fastened to a small rotating platform, so that the slit angle could be set to $\alpha_{slit} = 42^\circ$, which deviates slightly from what was determined by Oskar Öjstedt. This was because the tilt was decided by placing three candles in the beam path to see where all three were sharply imaged, which was best estimated at $\alpha_{slit} = 42^\circ$. The whole slit construction was ultimately mounted on the dovetail optical rail just as the rest of the receiver optics. The slit construction can be seen in Figure 3.3 and its placement on the dovetail optical rail in Figure 3.2.

The last part of the experimental setup was placing the detector. The detector was placed on a small platform that in turn was mounted on a rotating platform. The detector angle was set to $\alpha_{det} = -22^\circ$ as was determined from Eq.(2.9). The rotating platform was mounted on a translation stage that could move the detector with micrometer precision along the optical axis. This choice was made so that the detector can be easily adjusted to be located at the image plane of the slit. The detector was also mounted on a height-adjustable platform. Figure 3.4 show the full detector setup.

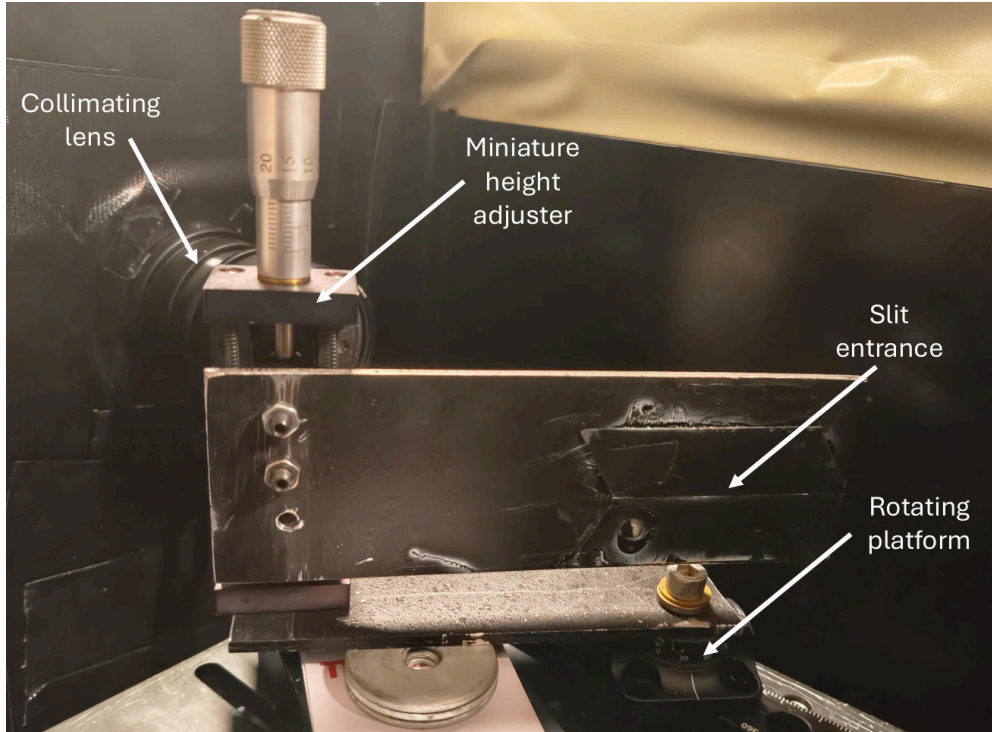


Figure 3.3: *The slit mount. Two box-cutter razor blades coated in black paint have been glued to a metal piece, which is fastened to a miniature height-adjuster. The platform the height-adjuster is fastened to is connected to a rotating platform mounted on the dovetail optical rail.*

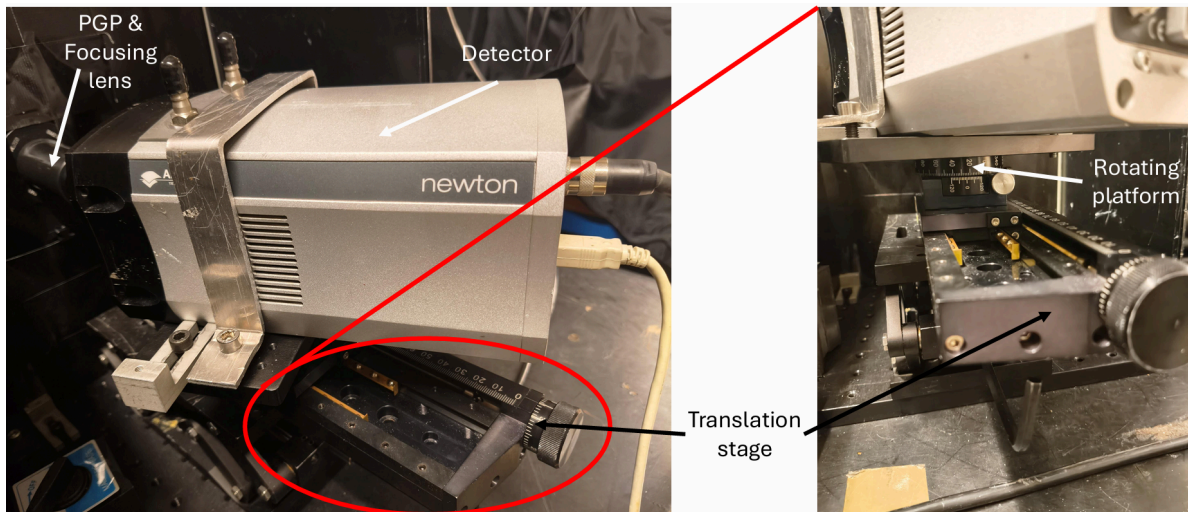


Figure 3.4: *The detector is placed on a rotating stage, a translation stage, and a height-adjustable platform.*

It is not, however, necessary to locate the vertical center of the detector on the optical axis. This is due to the PGP dispersing light vertically as demonstrated in Figure 2.8. By adjusting the detector's height, the wavelength interval it sees can be adjusted, as illustrated in Figure 3.5. This can also be understood by Figure 2.9, where if the rainbow image stays in place, the detector grid can be moved up and down to see different wavelength ranges. A notch filter and a long-pass filter centered at the laser wavelength set a minimum value that serves as a signature to guide the determination of the detector height.

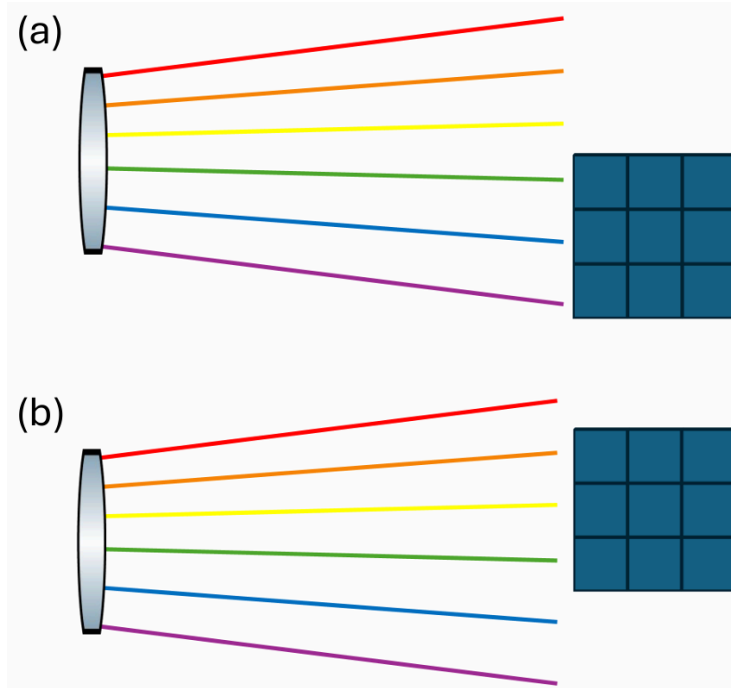


Figure 3.5: *The side view of the focusing lens focusing different wavelengths at different heights. (a) Shows the detector in one position as it sees one wavelength interval. (b) Shows the detector at a higher position, thus seeing a different wavelength interval.*

3.3 Terminal

The lidar setup allows for measuring ranges longer than a lab environment can accommodate. Thus, a thorough terminal needed to be constructed in the lab, mainly for safety purposes, but also to reduce the interference from other sources, even if small. A simple beam dump is not enough, as light still scatters from it. The terminal in Figure 3.6 was constructed to limit unnecessary interference as much as possible. It used a mirror to reflect the laser beam into a small enclosure surrounded by black-coated barriers. A black tarp was laid over the whole terminal to further limit the scattering. Initially, the beam dump was located where the mirror is, but it turned out that the scattered light still managed to exit through the small opening. Thus, the mirror was added to redirect the beam to an area where scattering would minimally exit through the opening.

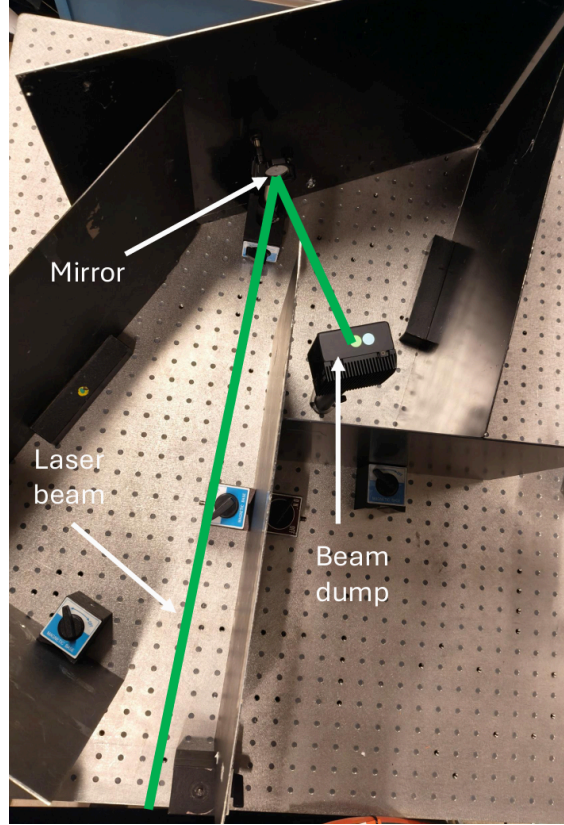


Figure 3.6: A mirror reflects the laser into a beam dump. This was surrounded by blockers to prevent any scattering. On top of the terminal was a black tarp for the same reason.

3.4 Alignment of the experimental construction

Firstly, every component of the receiver optics was placed on the dovetail optical rail with the relative distances between each component outlined by Oskar Öjstedt [16]. Since the optical rail acts as a ruler, this was simple. The construction of the slit did not allow for it to change position once mounted. For this reason, the positions of all other components were fixed relative to the slit. This was slightly difficult because the receiver lens and collimating lens were inside lens tubes, and the optical rail markings did not help to determine the precise position of the lenses. The receiver lens, slit, and collimating lens were adjusted as best possible to fit the optimized distances given by Oskar Öjstedt.

The distance d between the transmitter and receiver lens was set to 200 mm at $\varphi = 0$. When rotated to $\varphi = 5.5^\circ$, d changed to 190 mm, which was an oversight not accounted for. The attached video objective can be used to adjust the vertical angle of the laser beam, aiming to keep it as horizontal as possible relative to the table it stands on. A deviation of a fraction of a degree would lead to a noticeable height difference above the optical axis at greater distances, and hence would not be imaged onto the slit entrance, but instead below it, which ideally has to be avoided.

When the laser beam was made as horizontal as possible, the height of the laser beam was matched with the optical axis of the receiver optics. The height of the slit was then adjusted so that the image of the laser beam passes through the slit entrance. It could then continue to the detector. A single candle was first placed in the beam path to verify that the Scheimpflug imaging technique worked. The laser was then turned on to image the oxygen, nitrogen, and water vapor peaks on the detector. The detector (both rotation and position) was then adjusted using the translation stage and rotation platform to find where all peaks were simultaneously sharply imaged. The translation stage was adjusted in increments of $100\mu\text{m}$. It was possible to achieve precision down to $5\mu\text{m}$, but this level was deemed unnecessary within the allocated time, as $100\mu\text{m}$ increments provided a satisfactory result. Images of the three ambient air peaks were recorded and saved at each detector position for qualitative comparisons. The position of the image plane was found when all three peaks were in focus. The detector could be placed at the image plane of the slit with $100\mu\text{m}$ precision. The side view of the final receiver optics from the receiver lens to the detector is displayed in Figure 3.7, just as mentioned in Figure 2.10, and the total setup with the transmitter is seen in Figure 3.8 with a top view and no barriers included.

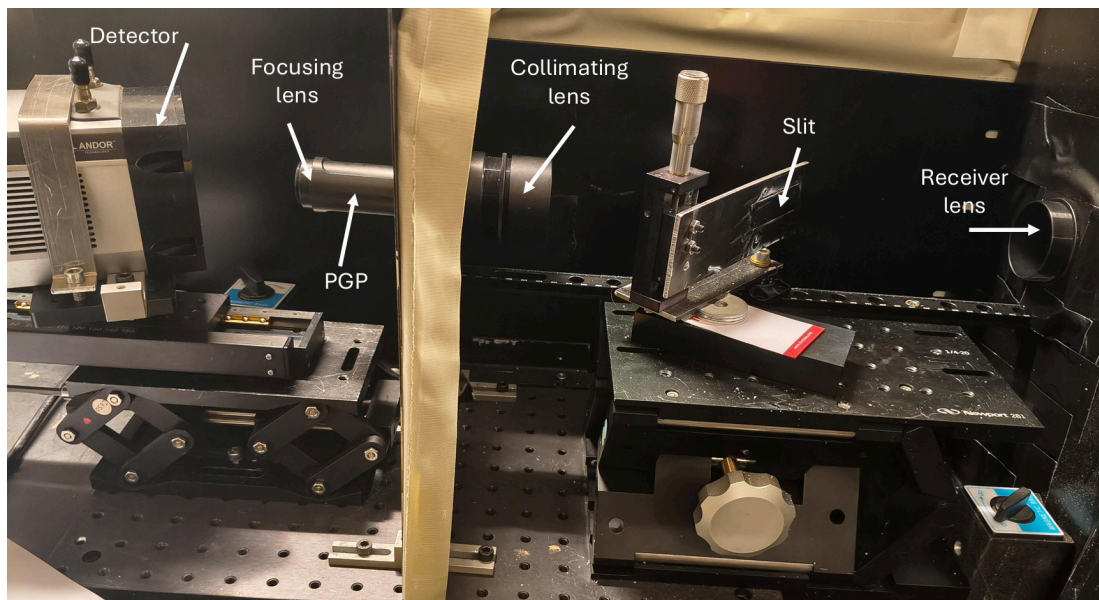


Figure 3.7: *Sideview of the complete setup of the receiver optics. The light enters the receiver lens to the right, passes through the slit, and then passes through the collimating lens. The PGP spectrally disperses the light, and the focusing lens images the light onto the detector.*

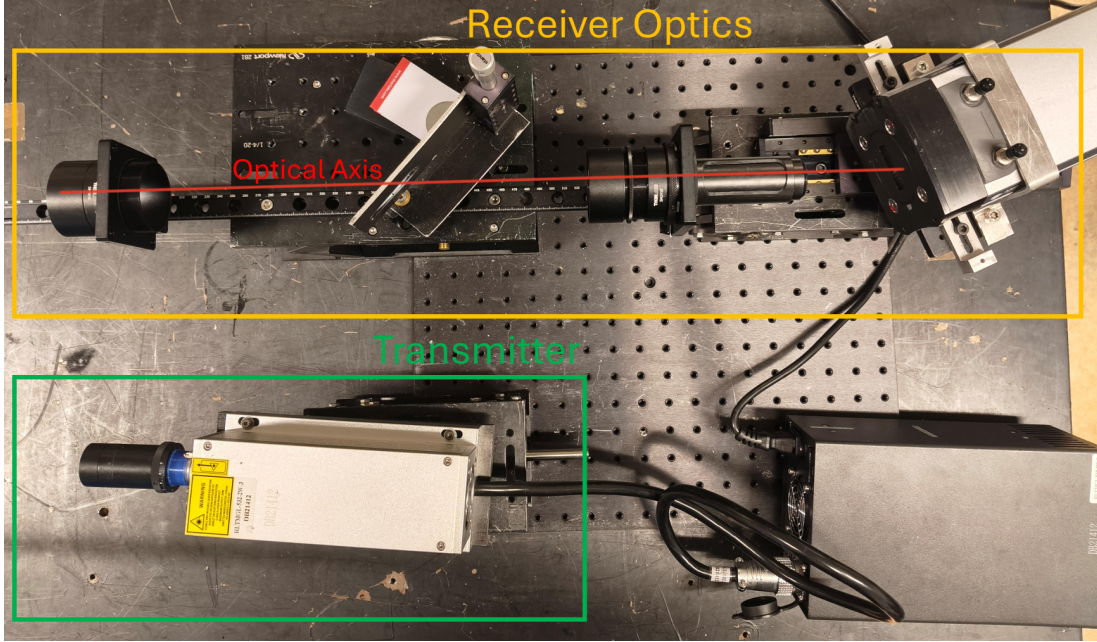


Figure 3.8: *Top view of the complete experimental setup, without barriers included. The transmitter has been marked in green, the receiver optics in orange, and the optical axis in red.*

Lastly, the detector height was adjusted using the height-adjustable platform so that the wavelength interval captured the Raman signal from soot up to water vapor at $\nu = 6345 \text{ cm}^{-1}$. The wavelength interval was decided by Oskar Öjstedt in his design of the PGP, which assumes the detector is vertically centered on the optical axis. The designed interval was 540-642 nm (corresponding to Raman shifts $278 \text{ cm}^{-1} - 3221 \text{ cm}^{-1}$), but it differed in practice when adjusting the detector height, as the said interval did not contain the water vapor peak at 660.37nm, even though it did in practice.

3.5 Calibration

3.5.1 Spatial calibration

Eq.(2.14) will be used to determine the distance to any source. All parameters except b are determined as part of the experimental construction. That is because b is not possible to measure with a ruler since, by its definition, it is the position on the slit which corresponds to $p_r = 0$, which cannot be determined manually. For this reason, a calibration point must be measured before measuring additional distances. A black barrier was placed in the beam path of the transmitter, and the distance, r_{ref} , was measured between the barrier and the transmitter (which is located on the Scheimpflug line). A barrier will scatter a lot, but for range calibration, this is fine since it does not care about wavelength. An image was recorded with the laser turned on at very low power. The image can then be summed over all wavelengths at each pixel column to get the total intensity. In a dark room, a sharp

intensity spike should be present at the pixel column that registers the barrier's image. Finding this pixel column $p_{r,ref}$ is the final step to determining b , since it can be solved for with the measured r_{ref} and $p_{r,ref}$.

3.5.2 Spectral calibration

With the experimental setup fully constructed to image the laser beam onto the detector, calibration data needed to be collected to determine which wavelengths each pixel corresponds to and thus determine the Raman shift using Eq.(2.2). By design, the wavelength range contains an oxygen peak at a Raman shift $\nu_{O_2} = 1555\text{cm}^{-1}$, which corresponds to a detectable wavelength at $\lambda_{O_2} = 580\text{nm}$ from a 532nm laser. Similarly, nitrogen and water vapor peaks with Raman shifts of $\nu_{N_2} = 2331\text{cm}^{-1}$ and $\nu_{H_2O} = 3654\text{cm}^{-1}$ respectively are detected at wavelengths $\lambda_{N_2} = 607\text{nm}$ and $\lambda_{H_2O} = 660\text{nm}$. These peaks are present in ambient air without any soot source present. With the laser on, the three peaks were identified on the detector image and were present across all detectable ranges, spanning many different pixel columns p_r . This provided more than a sufficient amount of data points covering a wide range of p_r and p_λ values at known wavelengths. These points were used to fit Eq.(2.15). Once the fitted parameters $a_0 - a_5$ had been determined, Eq.(2.15) was used to determine the wavelength of any pixel across the full detector grid.

3.6 Soot measurements

With the full experimental setup complete and calibrations performed, it was ready for soot measurements. Two types of measurements were made. The first type was a single, direct measurement made on different types of soot generated by a mini-CAST soot generator. The second type was a collection of measurements from the same source at different polarization angles.

3.6.1 Single direct measurement

The main purpose of the direct measurements was to preliminarily test whether a Raman signal from soot could be resolved using the Scheimpflug lidar configuration. A mini-CAST soot generator was placed roughly 2 meters from the transmitter, capable of generating a variety of soots. The two types of soot that were examined were black and brown soot. Black soot is generated at Operational Point 1 of the mini-CAST, henceforth referred to as OP1. Similarly, brown soot is produced at Operational Point 6, which is going to be referred to as OP6.

A long exposure time was necessary because of the weak Raman signal. A total exposure time was set to 1'000 seconds, split into 20 accumulations of 50 seconds each. A dark room measurement was first collected with the laser, and the mini-CAST turned off. This dark room measurement was subtracted from any subsequent measurements to remove electronic noise. With data collected to remove electronic noise and to both spatially

and spectrally calibrate any spectrum, the mini-CAST soot generator was set to OP1 to produce black soot, and a single measurement was recorded. Once that was done, the mini-CAST was set to OP6 to generate brown soot, and a single measurement was also recorded. This concluded the direct measurements.

3.6.2 Separation of polarized air contributions from the Raman signal

Since the Raman signal from soot overlaps with oxygen and carbon dioxide, they needed to be removed as best as possible. It was done by following a similar approach to that of Nilsson et. al. [29], who separated the polarized and unpolarized parts of a spectrum using Fourier analysis. This project will do the same to isolate the Raman spectrum of soot. This can be done because the Raman signal of soot is unpolarized [30], and the molecules in air and other gases are highly polarized [31], meaning the Fourier analysis could be done to separate them.

A polarizing beam splitter was placed immediately in front of the transmitter to obtain a linear polarization. A half-wave plate was placed directly after the polarizing beam splitter to rotate the laser beam's polarization angle. Both of these additions are seen in Figure 3.9.

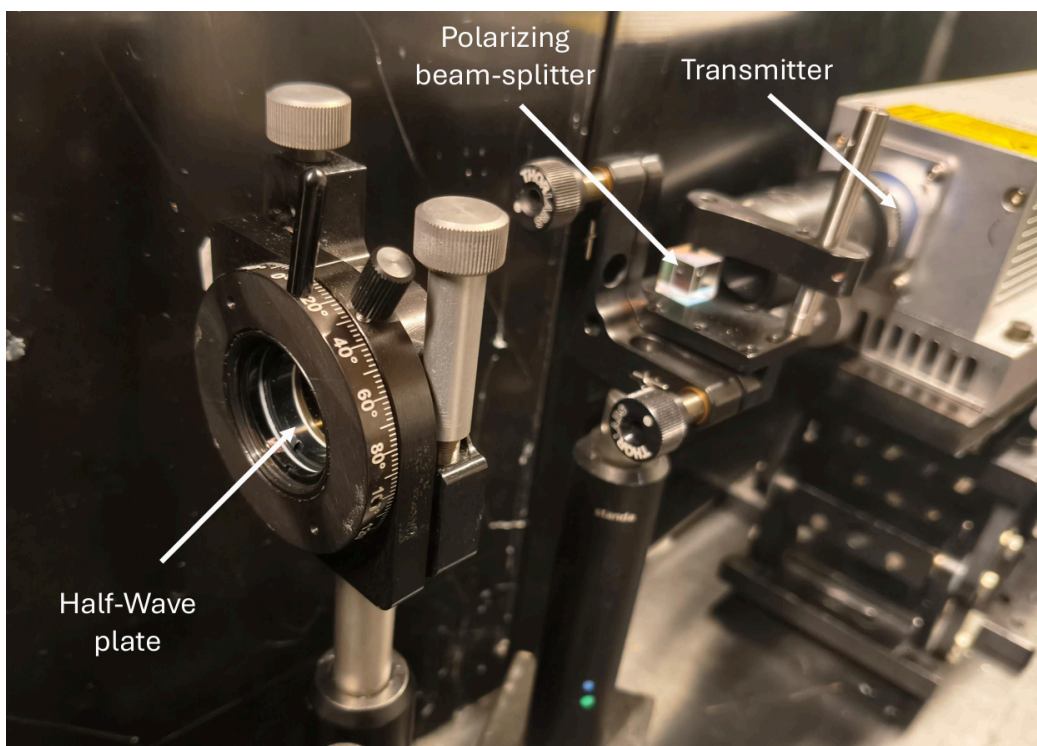


Figure 3.9: A beam-splitting polarizer and a half-wave plate have been introduced in the beam path right in front of the transmitter.

Finally, a polarizer was placed directly in front of the receiver lens and set to a fixed, arbitrary angle. Since oxygen, nitrogen, and water vapor are highly polarized, their intensities on the detector will oscillate as the polarization angle of the laser beam is rotated by the half-wave plate. The polarization angle needed to be rotated by rotating one full period, i.e., 360° , which was achieved by rotating the half-wave plate by 180° . The step size for each measurement was $\Delta\theta = 10^\circ$ for the half-wave plate, and thus $\Delta\theta = 20^\circ$ for the laser polarization. Eq.(2.18) tells that a total of 18 measurements should be recorded for each type of soot, alongside 3 measurements for dark noise, spectral calibration, and spatial calibration. All measurements, except the spatial calibration, are set to 1'000 sets of total acquisition time, resulting in a total measurement time of approximately 5.5 hours per operational point. Due to this long measurement time and the sensitivity of the experimental setup, it had to be done extremely carefully, yet it was necessary to do it in one go without interruption.

After spectral calibration, the intensity at the pixel column of the mini-CAST $p_{r,mini-CAST}$ can be described as $I_{mini-CAST}(\nu, \theta_n)$. Using Eq.(2.17), this became $\tilde{I}_{mini-CAST}(\nu, k)$, after being normalized using Eq.(2.19). From then, the DC component $I_{mini-CAST,DC}(\nu)$ was extracted using Eq.(2.23) after k_{AC} had been identified. If done correctly, the plot of $I_{mini-CAST,DC}(\nu)$ should be a close replica of the single direct measurement during data collection when the polarization angle of the laser beam was perpendicular to the polarizer angle. The reason behind this can be understood by noticing in Figure 2.12 that $f_{DC}(x)$ is located at the minimum of the oscillation.

4 Results

4.1 Construction and optimization of the experimental setup

After the setup was constructed but not optimized, a candle was placed within the laser beam path to see whether it indeed passed through the slit and was properly imaged onto the detector. Which it did as shown in Figure 4.1 as the red vertical line to the right. This confirmed the initial rough construction of the setup, and could move on to be optimized.

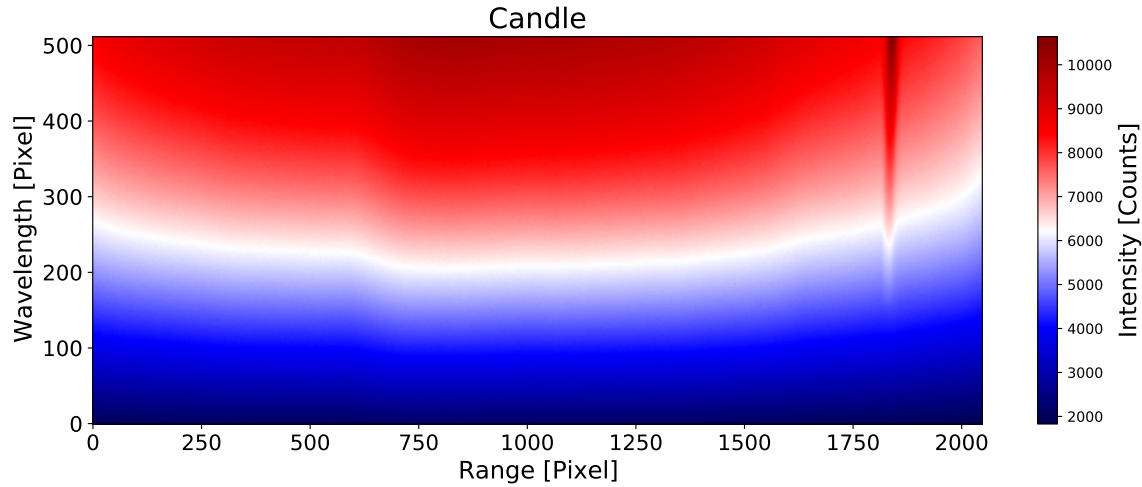


Figure 4.1: *A test candle was placed in the beam path to test that it could be mapped through the optics to the detector.*

The first challenge was to make the laser beam horizontal with respect to the table, which proved difficult due to the video objective. The laser itself did not produce a perfectly horizontal beam, but the video objective attached to it could change the direction. Adjusting the video objective is a very clunky way to fix the laser beam, but no other method was thought of. Ultimately, it was with the help of multiple irises, placed with their openings aligned as horizontally as possible, that the laser path was best horizontally adjusted. Given the significant imprecision of this method, the ultimate orientation of the laser beam was not perfectly horizontal, but it was sufficient to continue the project. It was then easy to adjust the height of the laser beam to align it with the optical axis of the receiver optics. It was done by using a ruler to measure the height from the table surface to the center of the receiver lens and marking it on the ruler. The optical axis was located approximately 20.4 cm above the table surface. The laser height was then adjusted so that the beam hit the mark on the ruler.

The slit needed more work to align with the optical axis. Because the image of the laser beam is relatively thin, almost as thin as a strain of hair, i.e., on the scale of micrometers, the slit needed extremely precise adjustments until the light from the laser beam image

passed through the slit. Even though the detector was not perfectly aligned, it could still register an image, as in Figure 4.1. If the laser is then turned on and the slit is aligned with the optical axis, the image will pass through the slit and be registered on the detector. Through a tedious process of changing the height of the slit in increments of $10\mu m$, and looking at the detector image to see whether the nitrogen peak was visible or not, the position of the slit was ultimately located as close to the optical axis as possible with around $10\mu m$ precision.

Now that the laser beam could be imaged onto the detector, the detector position was optimized. It was adjusted with $100\mu m$ precision using a translation stage over a range of 2 mm, at angles of $\alpha_{det} = 20^\circ, 22^\circ, 23^\circ, 24^\circ, 25^\circ$. At each position, a spectrum was recorded and saved. Each image showed three peaks across a range of wavelengths: oxygen, nitrogen, and water vapor. By finding the peak amplitudes and plotting them as a function of the distance between the focusing lens and detector, the image plane was found to be best at the angle and position when all three species had their absolute maxima at the same position. This came out to be at $\alpha_{det} = 22^\circ$ as expected at a distance between the focusing lens at detector of 73 mm. With the construction of the setup optimized within the limits of what was available and within the time constraint, it could be used to do measurements on soot generated by a mini-CAST soot generator.

The construction of the setup followed Oskar Öjstedt's values as closely as possible. Table 4.1 highlights the values given by him, and the values put into practice after optimization. The tilt of the slit and detector in the simulation were $\alpha_{slit} = 43.2^\circ$ and $\alpha_{det} = 33.3^\circ$ respectively. Meanwhile in the experiment the values were $\alpha_{slit} = 42^\circ$ and $\alpha_{det} = 22^\circ$.

Table 4.1: *The distance between components from the simulated setup compared with the real experimental setup.*

	Simulation [mm]	Reality [mm]
Receiver lens to slit	211	211
Slit to collimating lens	141.2	142
Collimating lens to iris	3	3
Iris to notch filter	3	3
Notch filter to first prism	5.5	5.5
First prism to grating	3	3
Grating to second prism	3	3
Second prism to long-pass filter	5.5	5.5
Long-pass filter to focusing lens	6	6
Focusing lens to detector	75.85	73

4.2 Direct measurements of soot

4.2.1 Spatial calibration

To obtain meaningful information from soot measurements, the detector needed to be calibrated. The first measurement was for spatial calibration. A black barrier was placed at a distance of $r_{ref} = 1606$ mm from the transmitter, and its image from the scattering of the laser at very low power was recorded in a single shot with an exposure time of 0.5 sec. Figure 4.2 shows this image, where the red vertical line to the right is the image of the scattering from the reference barrier.

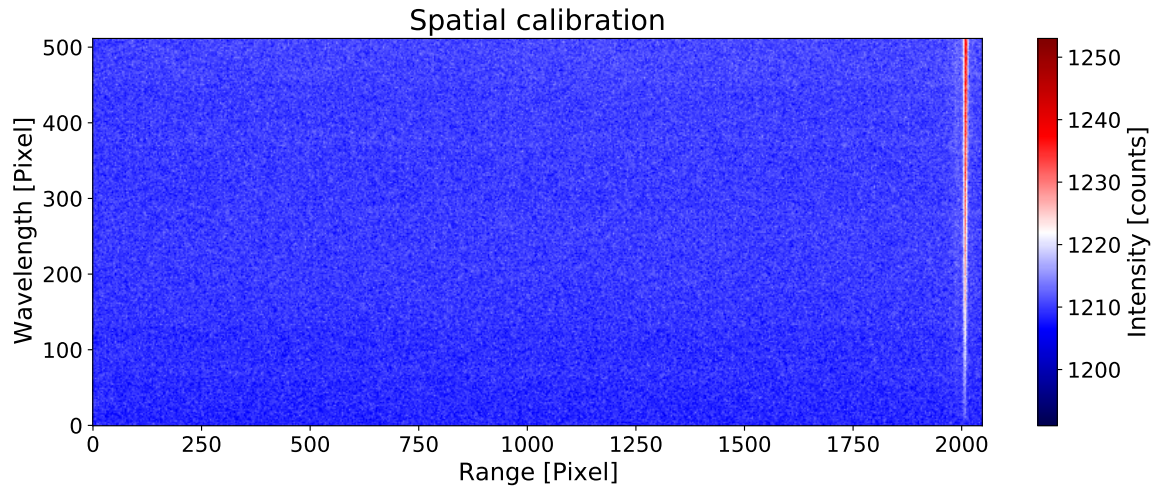


Figure 4.2: Image of a black barrier placed at a reference distance of $r_{ref} = 1606$ mm away from the transmitter. The pixel column for the barrier is $p_{r,ref}$, which will be used for spatial calibration.

This spectrum was then summed over all wavelengths to obtain the total intensity for each pixel column. Figure 4.3 shows a zoomed-in image of this around the location of the beam dump. Here, the pixel column with the highest total intensity was identified as $p_{r,ref} = 2007$. With the help of Eq.(2.14), the parameter b was determined to be $b = 194.6$ mm. A quick sanity check tells us this is expected since b should be within the slit range. The distance between the receiver lens and the slit along the optical axis is 211 mm, as outlined by Oskar Öjstedt, meaning the value of b per its definition illustrated in Figure 2.10 cannot be larger than that as otherwise the detector would not cross the optical axis at all. The width of the slit is $l_{slit} = 50$ mm, half of which is on each side of the optical axis, and its tilt is $\alpha = 42^\circ$, meaning the projection of the slits lower end on the optical axis is a distance of 194.3 mm away from the receiver lens. A value of $b = 194.6$ mm falls within the expected range and is very close to the very end of the slit, indicating the detector is well horizontally centered, but not perfectly so.

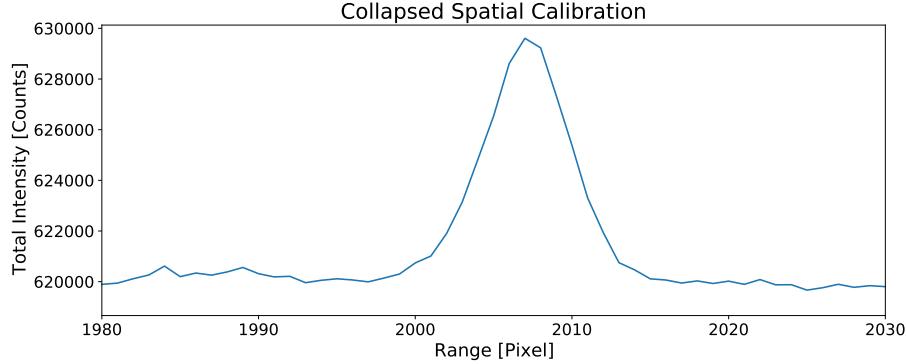


Figure 4.3: Total intensity along pixel columns around the reference beam dump. The peak is identified as the beam dump, and the reference pixel $p_{r,ref}$ is chosen as the column with the largest total intensity from the scattering.

4.2.2 Spectral calibration

Now that the detector had been spatially calibrated, the next step was spectral calibration. Since some molecules in ambient air are prominent in the close vicinity of the Raman spectrum of soot, they will be used for spectral calibration. To achieve the clearest possible distinction between the peaks, the measurement was taken in a completely dark room with only the laser turned on. The receiver optics were also isolated as best as possible using barricades to limit scattering from the surroundings. The detector collected data for 20 accumulations at 50 seconds each, for a total exposure time of 1'000 seconds. The resulting spectrum in Figure 4.4 shows three lines, each corresponding to three different molecules present in ambient air. The bottom line is oxygen centered at 1555 cm^{-1} , the middle line is nitrogen at 2331 cm^{-1} , and the top and faintest line is water vapor at 3654 cm^{-1} .

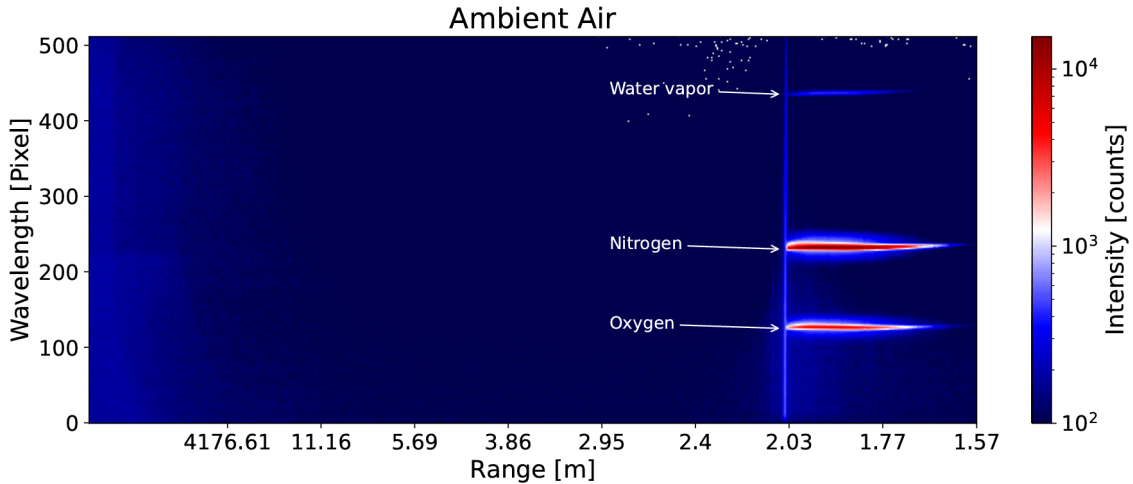


Figure 4.4: A measurement with no soot source present within the beam path. The three peaks from bottom to top are identified as oxygen, nitrogen, and water vapor.

A total of 280 pixel columns containing all three peaks was selected. Since each column has three data points, a total of 840 data points were picked. Each point in this data set has a known Raman shift, pixel column p_r , and pixel row p_λ . The Raman shifts were converted to wavelength using Eq.(2.2), and then all data points were used to best fit the spectral calibration function in Eq.(2.15) to get the calibration parameters $a_0 - a_5$, which are presented in Table 4.2.

Table 4.2: *Spectral calibration parameters $a_0 - a_5$ as best fitted using 840 data points.*

a_0	543.8 ± 0.6
a_1	0.280 ± 0.005
a_2	$(22 \pm 3) \cdot 10^{-4}$
a_3	$(4 \pm 8) \cdot 10^{-6}$
a_4	$(-1.3 \pm 0.3) \cdot 10^{-5}$
a_5	$(0.8 \pm 5) \cdot 10^{-9}$

One thing to note is that the uncertainties for a_3 and a_5 are larger than the fitted value. The reason can be explained by either an incorrect calibration function or imprecise data. In reality, the slit should let light pass through over the whole range from closest to the terminal. Figure 4.4 suggests that the slit entrance is not completely horizontal. If it were, the horizontal lines would extend to the edge at $p_r = 2048$, which they do not. This suggests the data may be slightly skewed. So a larger data set and a sharper image should yield better-fitting parameters. The issue was not attempted to be remedied due to the limited time available for the project, and their contributions were close to negligible.

4.2.3 OP1 soot measurements

Before any soot measurements were taken, a dark noise measurement was taken. This was with the same 1'000 total exposure time as before in a completely dark room, meaning no lights and no laser turned on. This dark noise measurement is later subtracted from the soot measurement to obtain only the signal from the laser beam.

The mini-CAST soot generator that produced the soot stream was placed approximately 2 meters away from the transmitter. The mini-CAST soot generator was then set to Operational Point 1, corresponding to the generation of black soot. The measurement was taken with a total exposure time of 1'000 seconds, and after dark noise removal, we obtain the image shown in Figure 4.5. The left-most vertical line that looks to block the horizontal lines is the terminal. The vertical line near the center of the plot is the signal from the soot. This proved that a 2W CW laser passing through soot does produce a detectable, distinguishable signal.

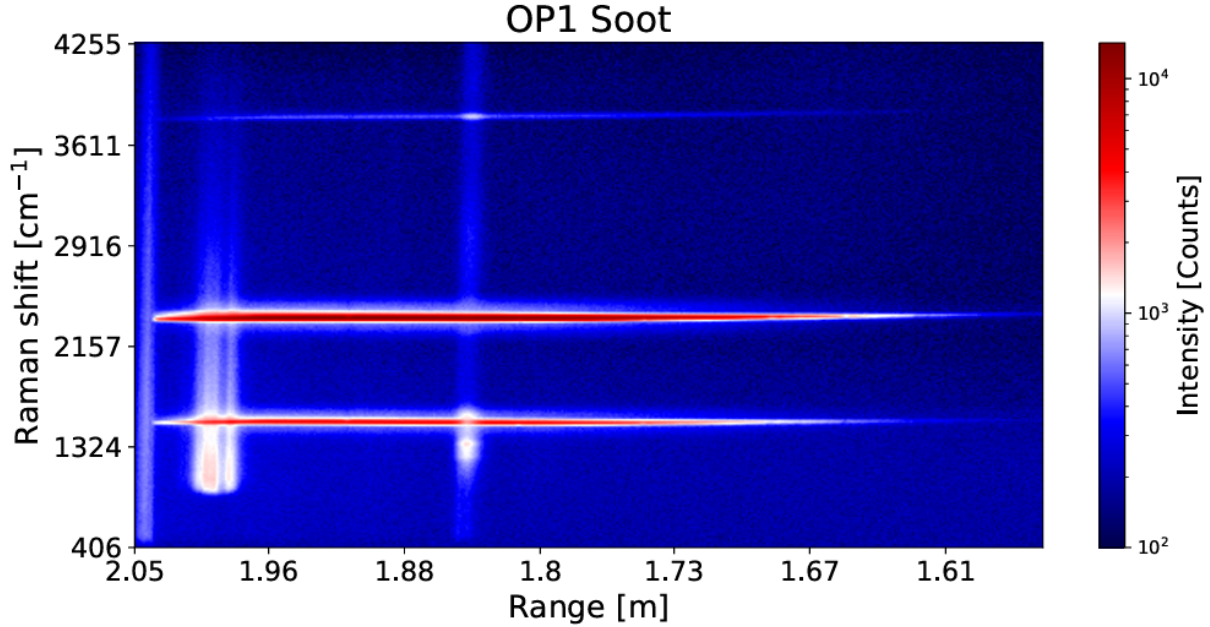


Figure 4.5: *Intensity heat map zoomed in to the range covered by the detector. The vertical line close to the center of the image is the signal from the black soot generated by the mini-CAST.*

The pixel column of the soot that has the strongest signal is at $p_r = 1769$. Since the pixel column covers distances between $p_r = 1768$ and $p_r = 1769$ as explained in Figure 2.11, the distance to the soot generated by the mini-CAST soot generator is determined to be between 1.8433 m and 1.8445 m. This deviates approximately 20 cm from the actual location. The reason for this deviation is that the experimental construction does not match the Scheimpflug theory outlined in Eq.(2.3) and Eq.(2.4), which is necessary for Eq.(2.14) to be accurate. For $d = 190$ mm and $\varphi = 5.5^\circ$, we would expect $S = 222.6$ mm and $\alpha = 49.5^\circ$. This deviates noticeably from the experimental setup, where $S = 211$ mm and $\alpha = 42^\circ$.

The spectral range came out to be 548-680 nm (534 - 4089 cm^{-1}), and the spectral resolution at short wavelengths was $\Delta\lambda = 5.7$ nm to $\Delta\lambda = 8.5$ nm at longer wavelengths. Since it was confirmed that a signal from the soot was detectable, the next step is to see whether it could resolve the D and G peaks of soot. The intensity spectrum at $p_r = 1769$ was selected and spectrally calibrated. This gave the x-axis in units nm. Eq.(2.2) was used to convert into ν_{Raman} so that the spectrum instead showed the Raman shift on the x-axis. The plot for this is shown in Figure 4.6. This spectrum shows, from right to left, a small water vapor peak, a large nitrogen peak, and, finally, the Raman signal from soot in the form of the D and G peaks.

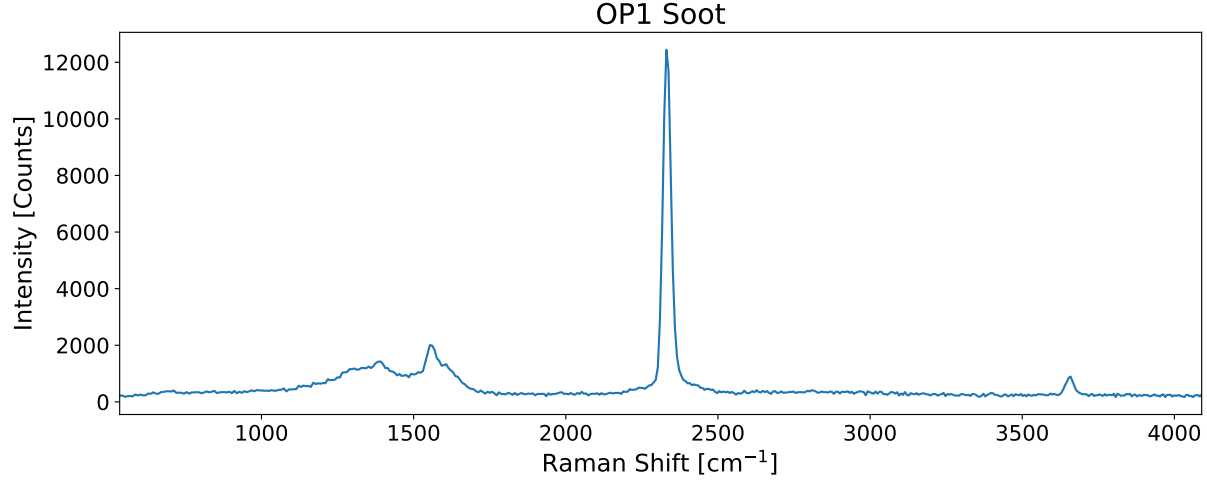


Figure 4.6: *Spectrally calibrated intensity spectrum of black soot generated by a mini-CAST approximately 2 meters away from the transmitter.*

This confirms that the Raman signal of soot can be spectrally resolved using the Scheimpflug configuration. Next, it was checked whether the spectrum was good enough to gather any information from. The resolution is worse than previous *ex-situ* measurements [11, 16]. For this reason, it was decided to only include the D , G , and D_3 peaks centered around 1360 cm^{-1} , 1600 cm^{-1} , and 1500 cm^{-1} , respectively. The D and G peaks were fitted using Lorentzian functions, while the D_3 peak and gas contributors such as oxygen and two carbon dioxide peaks were fitted using Gaussian functions. The fluorescent background was removed using a linear fit of the form $mx + b$. The spectrum in Figure 4.7 shows the deconvolution of the spectrum after the fluorescent background has been removed.

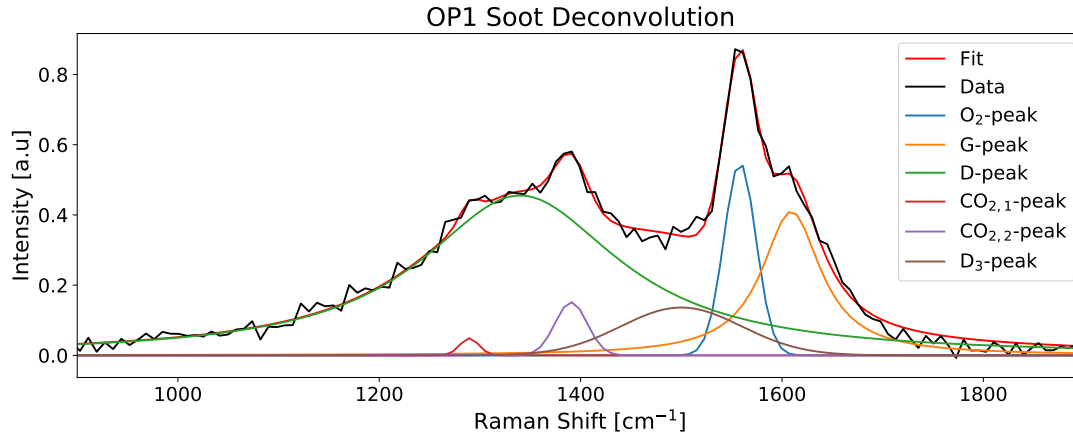


Figure 4.7: *The deconvolution of the OP1 soot spectrum generated by the mini-CAST.*

The relative intensity, I , peak center, x_0 , and the FWHM, Γ , of each peak are presented in Table 4.3. The linear fluorescent background that was removed had a slope of $m = (-22 \pm 4) \cdot 10^{-6}$ and a y-axis intercept of $b = 0.162 \pm 0.007$.

Peak	I [a.u]	x_0 [cm $^{-1}$]	Γ [cm $^{-1}$]
$CO_{2,1}$	0.05 ± 0.02	1289 ± 4	22 ± 11
D	0.46 ± 0.02	1340 ± 10	240 ± 16
$CO_{2,2}$	0.15 ± 0.02	1391 ± 2	37 ± 6
D_3	0.14 ± 0.03	1500 ± 20	120 ± 49
O_2	0.55 ± 0.05	1558.2 ± 0.7	38 ± 3
G	0.41 ± 0.05	1609 ± 2	71 ± 5

Table 4.3: Fitted values for peaks used in the deconvolution of OP1 soot. I is the peak amplitude, x_0 is the peak center and Γ is the FWHM.

4.2.4 OP6 soot measurements

The same data analysis was performed on OP6 soot (i.e., brown soot) as on OP1 soot. Dark noise, spatial and spectral calibration spectra used for OP6 analysis were the same as those used in the OP1 analysis, meaning only a new spectrum for OP6 soot had to be measured by setting the mini-CAST to OP6 to generate brown soot. The spectrum after dark noise subtraction is shown in Figure 4.8, where the vertical line of the soot is clearer due to the larger organic compound relative to OP1 soot and hence exhibits stronger fluorescence.

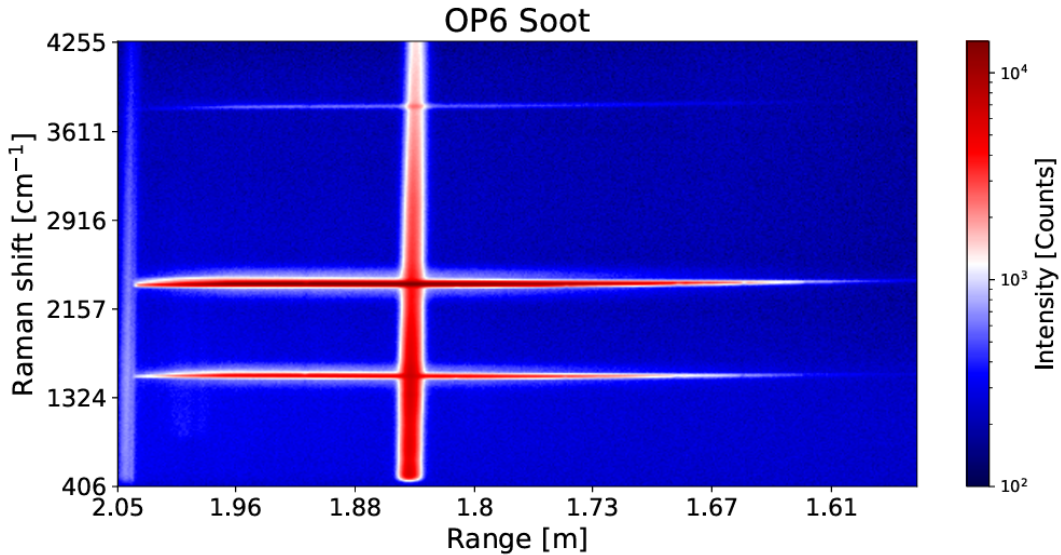


Figure 4.8: Intensity heat map zoomed in to the range covered by the detector. The vertical line close to the center of the image is the signal from the brown soot generated by the mini-CAST.

The spectrum at the same pixel column as for OP1 soot, at $p_r = 1769$, was retrieved and is shown in Figure 4.9, where the large fluorescence is clearly visible.

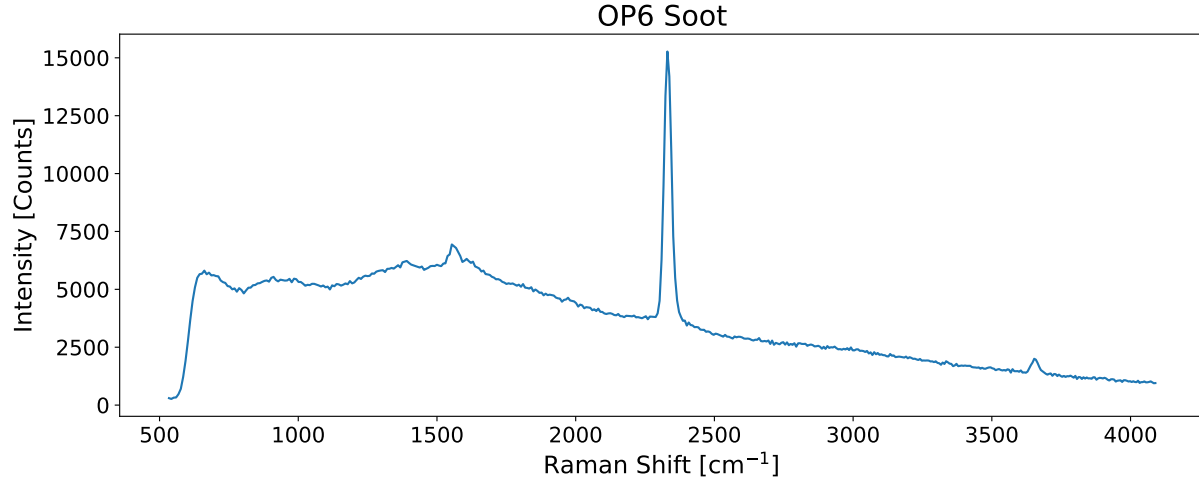


Figure 4.9: *Spectrally calibrated intensity spectrum of brown soot generated by a mini-CAST approximately 2 meters away from the transmitter.*

It is possible to see, even if ever so slightly, the Raman spectrum of soot, demonstrating that soot spectra can be resolved using the Scheimpflug lidar setup even in the presence of large fluorescence. The fluorescence was removed using a quadratic fit $ax^2 + bx + c$ with $a = (-53 \pm 2) \cdot 10^{-8}$, $b = (157 \pm 6) \cdot 10^{-5}$, and $c = -0.38 \pm 0.05$. The Raman spectrum of soot after removal of the fluorescent background is shown in Figure 4.10.

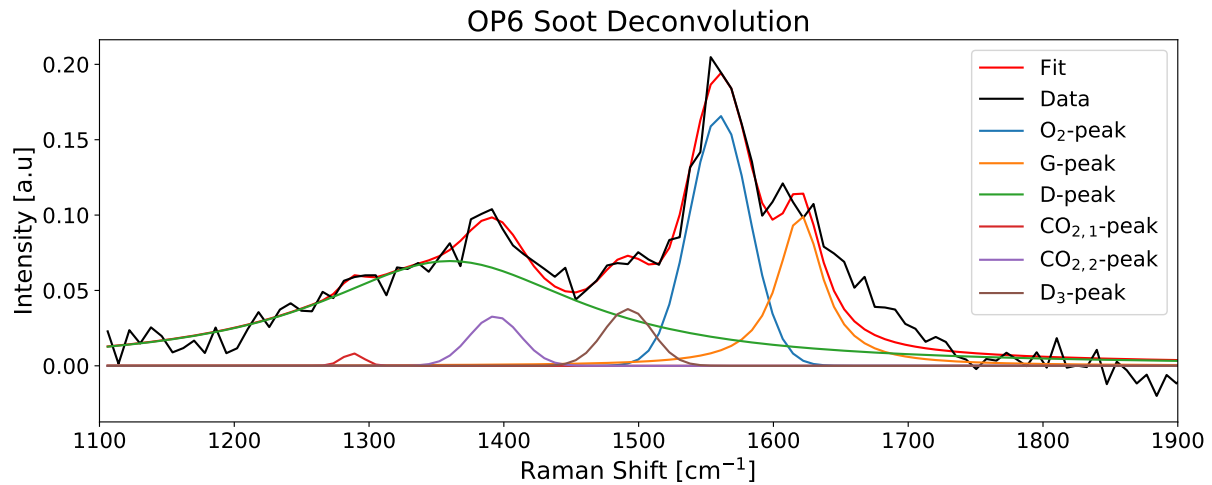


Figure 4.10: *The deconvolution of the OP6 soot spectrum generated by the mini-CAST.*

This spectrum also includes the fitted peaks, whose parameters are presented in Table 4.4. The fluorescent background relative Raman signal can now be compared between black an

Table 4.4: *Fitted values for peaks used in the deconvolution of OP6 soot.*

Peak	I [a.u]	x_0 [cm ⁻¹]	Γ [cm ⁻¹]
$CO_{2,1}$	0.008 ± 0.012	1288 ± 13	16 ± 29
D	0.07 ± 0.01	1360 ± 20	240 ± 65
$CO_{2,2}$	0.03 ± 0.01	1393 ± 6	38 ± 17
D_3	0.04 ± 0.01	1492 ± 6	36 ± 15
O_2	0.166 ± 0.009	1560 ± 1	52 ± 5
G	0.1 ± 0.01	1620 ± 3	40 ± 8

brown soot. Their respective plots using the fitted parameters were integrated and then summed to get their total area. The respective signals were then divided by the total area to get the percentage, the result of which can be seen in Figure 4.11.

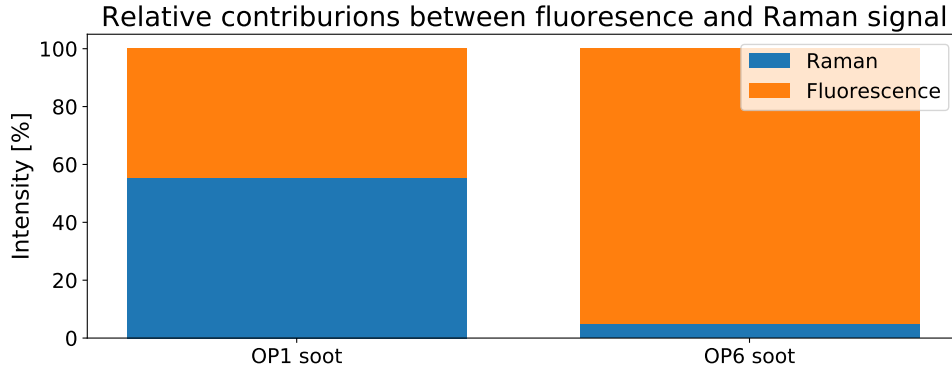


Figure 4.11: *A histogram showing the percentage contribution of fluorescent background and Raman signal using fitted parameters, for OP1 soot and OP6 soot respectively.*

4.3 Polarization measurements of soot

Since the direct measurements proved fruitful, both OP1 soot and OP6 soot can spectrally resolve the D and G peaks of soot, identify the fluorescent background, and retrieve information about the peaks that the spectra are composed of. This gave the green light to proceed to the next step: separating the polarized components, carbon dioxide and oxygen, from the Raman spectra of soot. The setup remained the same as for the direct measurements, and spatial and spectral calibration data were again collected. A beam-splitting polarizer and a half-wave plate were added in front of the transmitter, and a polarizer in front of the receiver lens. The polarizer was set to an arbitrary position and remained fixed. The beam-splitting polarizer kept the laser beam horizontally polarized, and the half-wave plate could rotate the polarization angle.

4.3.1 OP1 soot measurements

After dark noise, spatial and spectral calibration data were collected again, the mini-CAST was set to OP1. With steps of $\Delta\theta = 10^\circ$ on the half-wave plate, a full rotation of the laser beam was gotten after $N = 18$ measurements. The same pixel column for the mini-CAST position as in the direct measurements ($p_r = 1769$) was then picked out for each recorded image. They were then used to derive the DC signal using Eq.(2.23). To demonstrate how this works, we look at the nitrogen peak. The intensity and FFT of the intensity are shown in Figure 4.12.

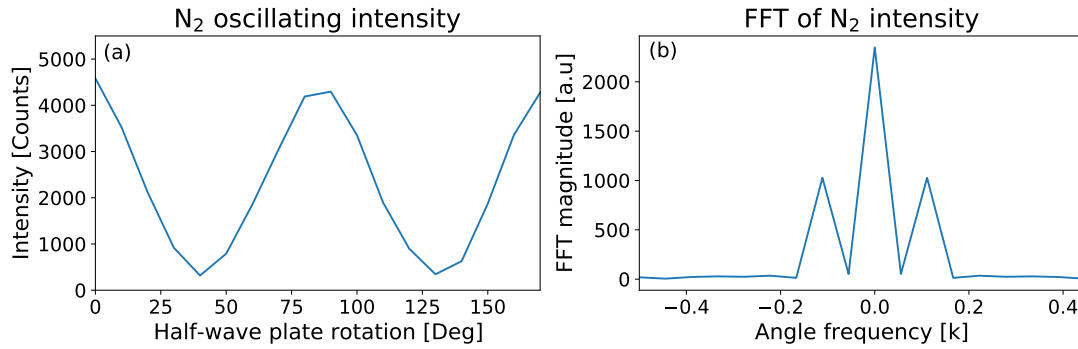


Figure 4.12: (a) The oscillating intensity of the nitrogen peak at $p_r = 1769$ as a function of the half-wave plate angle. (b) The FFT of the nitrogen signal has three clear spikes. The central spike is the average, and the two smaller spikes are the oscillation's amplitudes.

From Figure 4.12b, the amplitude at of the central spike at $k = 0$ is $\tilde{I}(\nu_{N_2}, 0) = 2347.4$ and the amplitude of one of the smaller spikes is $\tilde{I}(\nu_{N_2}, k_{AC}) = 1027.1$. From Eq.(2.23), this gave a signal of $I_{DC}(\nu_{N_2}) = 293.1$ which is about 6.7% of the maximum N_2 intensity. This procedure was done numerically for each ν within the spectrum, and Figure 4.13 shows the resulting spectrum of just the DC signal of OP1 soot.

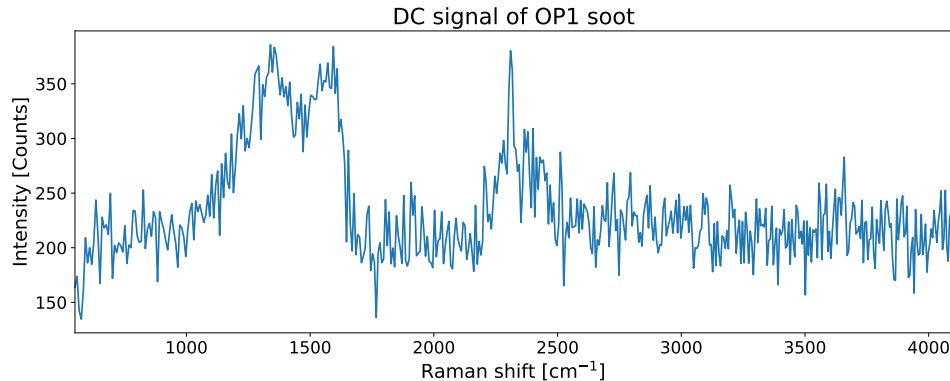


Figure 4.13: The spectrum of OP1 soot with polarized contributions minimized using Fourier analysis.

The AC signal was then separately plotted in Figure 4.14 to see which polarized gases and their respective intensities that were removed.

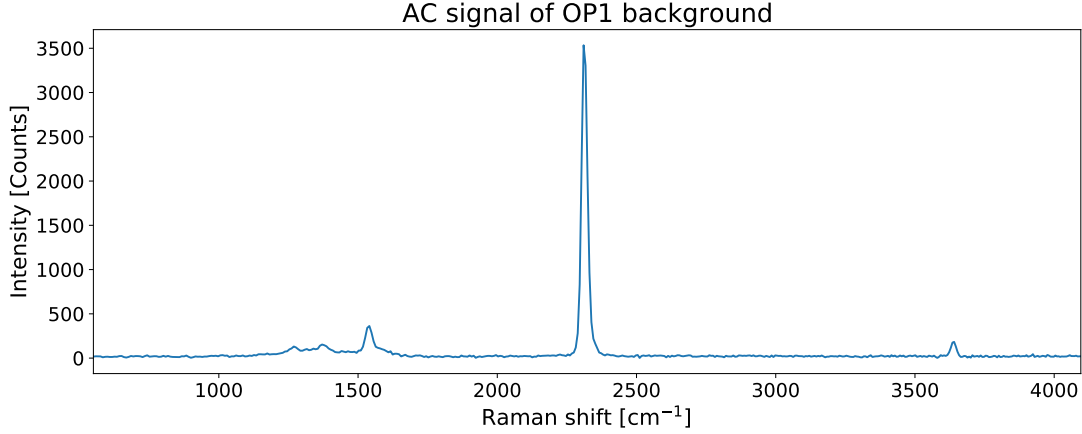


Figure 4.14: *The spectrum of the removed polarized contributions from OP1 soot.*

This spectrum shows that carbon dioxide and oxygen were removed from the OP1 soot spectrum, and they were not part of the final analysis. Considering this, the spectrum in Figure 4.13 underwent the same deconvolution as the direct measurement, which we can see in Figure 4.15. A linear fluorescent background with $m = (-1.7 \pm 1.0) \cdot 10^{-5}$ and $b = 0.54 \pm 0.02$ was removed from this spectrum.

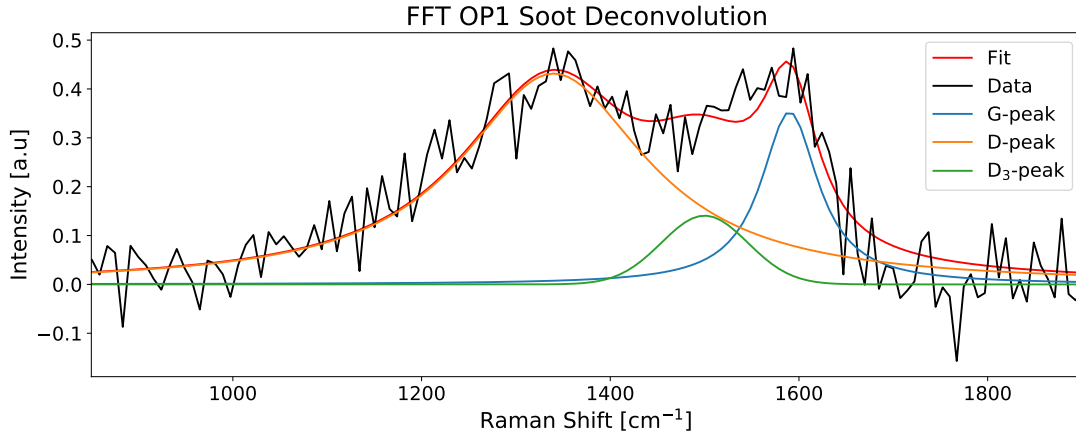


Figure 4.15: *The deconvolution of the DC signal of OP1 soot after Fourier analysis.*

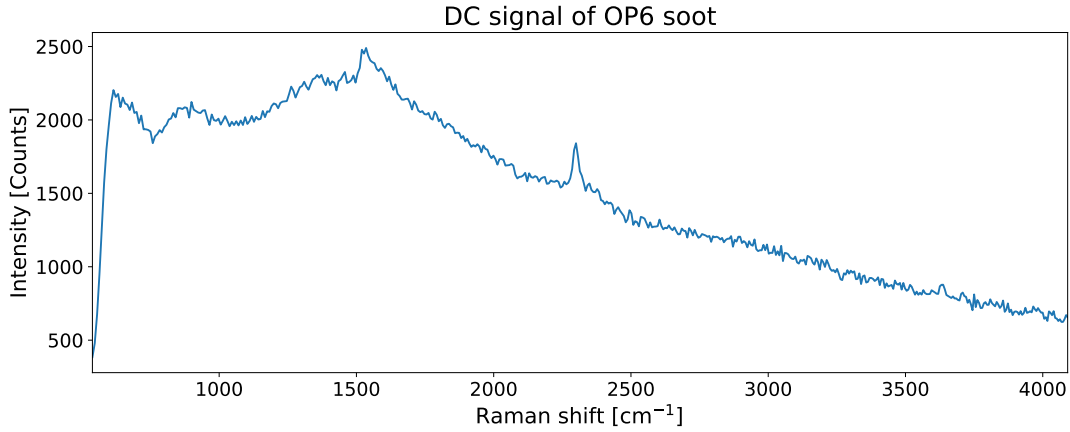
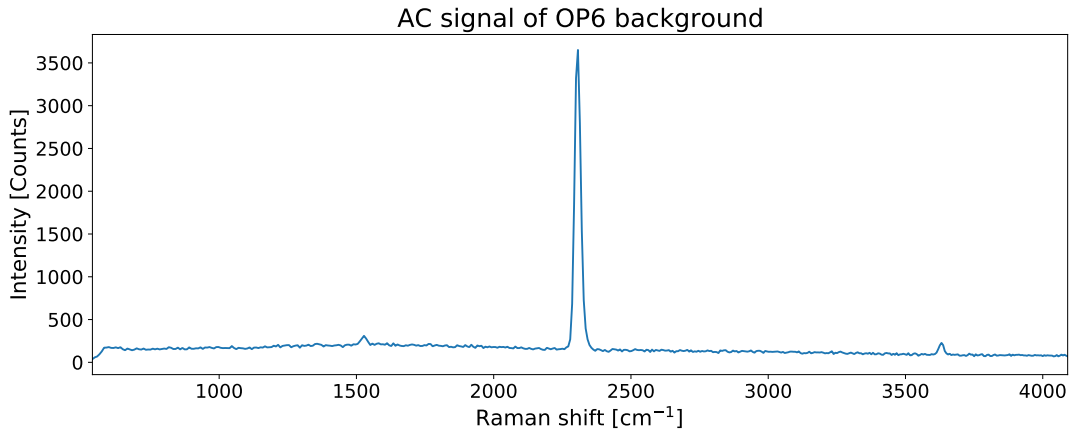
The fitted values from the deconvolution of the remaining spectrum are presented in Table 4.5

Table 4.5: *Fitted values for peaks used in the deconvolution of the DC signal of OP1 soot.*

Peak	I [a.u]	x_0 [cm^{-1}]	Γ [cm^{-1}]
D	0.43 ± 0.02	1340 ± 10	240 ± 29
D_3	0.14 ± 0.04	1500 ± 17	91 ± 40
G	0.35 ± 0.05	1590 ± 6	74 ± 13

4.3.2 OP6 soot measurements

The same procedure as was done for the Fourier analysis of OP6 soot as was done on OP1 soot at the same pixel column $p_r = 1769$. The DC and AC signals after separation are shown in Figure 4.16 and Figure 4.17, respectively.

Figure 4.16: *The spectrum of OP6 soot with polarized contributions minimized using Fourier analysis.*Figure 4.17: *The spectrum of the removed polarized contributions from OP6 soot.*

Since Figure 4.17 does not show that carbon dioxide has been removed, it was still part of the analysis. Likewise, for oxygen, it was deemed that its reduction was insufficient to warrant removal from the analysis. Figure 4.18 shows the deconvolution of the OP6 soot after Fourier analysis. A quadratic fluorescent background with $a = (-49 \pm 2) \cdot 10^{-8}$, $b = (144 \pm 6) \cdot 10^{-5}$, and $c = -0.21 \pm 0.05$ was removed. The fitted parameters are presented in Table 4.6.

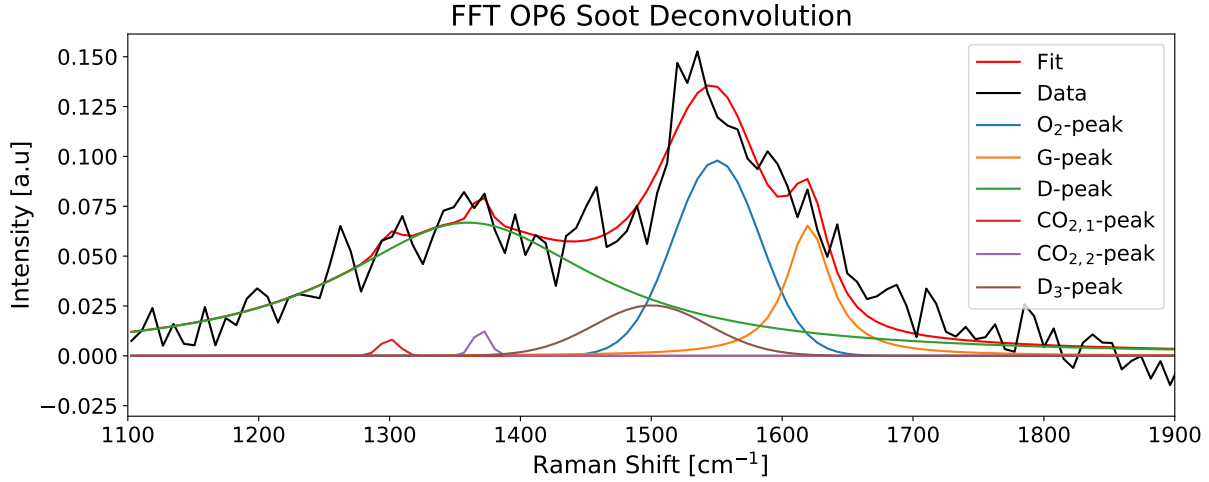


Figure 4.18: *The deconvolution of the DC signal of OP6 soot after Fourier analysis.*

Table 4.6: *Fitted values for peaks used in the deconvolution of the DC signal of OP6 soot.*

Peak	I [a.u.]	x_0 [cm ⁻¹]	Γ [cm ⁻¹]
$CO_{2,1}$	0.008 ± 0.013	1300 ± 13	18 ± 35
D	0.07 ± 0.02	1360 ± 55	240 ± 133
$CO_{2,2}$	0.013 ± 0.015	1370 ± 8	15 ± 23
D_3	0.02 ± 0.36	1500 ± 798	87 ± 699
O_2	0.10 ± 0.58	1550 ± 53	78 ± 82
G	0.06 ± 0.02	1620 ± 5	40 ± 14

5 Discussion

5.1 Experimental setup

The setup outlined by Oskar Öjstedt worked well with small adjustments made for practical reasons. The final mapping of distance r to pixel column p_r is seen in Figure 5.1, where a clear inverse relationship is made very prominent.

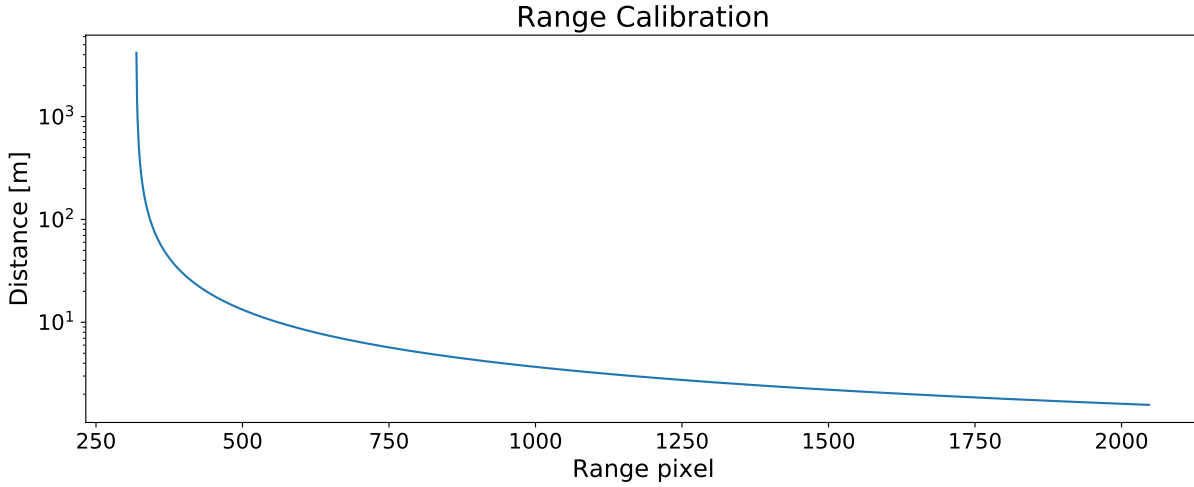


Figure 5.1: *The mapping of range measured by the SRL to the pixel columns p_r on the detector after calibration.*

The mini-CAST soot generator was measured with a measuring tape to be 2 meters from the transmitter, whereas the spatially calibrated spectrum yielded a range of 1.8433–1.8445 m. This discrepancy can be explained by the fact that Eq.(2.14) relies on the fact that Eq.(2.3) and Eq.(2.4) are fulfilled. As previously mentioned, these equations were not satisfied in Oskar’s setup. Similarly, for the magnification factor M_{4f} defined by Eq.(2.7), the distance between the object and collimating lens has to be equal to the focal length of the collimating lens, the distance between the collimating lens and focusing lens has to be equal to the sum of their focal lengths, and the distance between the focusing lens and the image has to be equal to the focal length of the focusing lens. It is these distances $f_{col} \rightarrow f_{col} + f_{foc} \rightarrow f_{foc}$ that define a 4f-optical system. These conditions were not fulfilled, meaning the magnification factor is also inaccurate. If ranging accuracy is valued for future constructions, it is vital that the conditions to use Eq.(2.14) are met.

Despite the inaccurate range determination due to the rough experimental setup, the method proved successful nonetheless. However, the slit construction, placement, and orientation can be improved for clearer images. The mount it is placed on can also be improved to better horizontally align it relative to the table it stands on. The mini height-adjuster shown in Figure 3.3 was too heavy for the platform it stood on, so it needed

support from below to prevent bending the platform and further misaligning the slit entrance.

To improve the overall system, the relative distances between components should better match the Scheimpflug theory and the requirements of the 4f optical system. The slit mount can be refined to better mount on the optical rail and have its entrance horizontal relative to the surface it stands on. The slit entrance can be made smaller to further reduce interference from external sources and scattering from the surroundings, resulting in a cleaner signal. The laser beam from the transmitter can also be made more horizontal relative to the table it is mounted on. For the placement of the slit, the detector can be placed where it should be and tested at different values of S and α to fully confirm which values give the sharpest focus, which should be that of Eq.(2.3) and Eq.(2.4). This can also test how well the laser's horizontal alignment is. The magnification due to the receiver lens, M_{rec} , can be derived from Eq.(B.2) and Eq.(B.3)

$$M_{rec} = -\frac{b + L' \sin(\alpha)}{r \cos(\varphi)} \propto \frac{1}{r} \quad . \quad (5.1)$$

Because of this inverse relationship, assuming a constant beam width perfectly centered on the optical axis, the image will look something like that in Figure 5.2 with arbitrarily set values, where the closest distance is on the left and experiences the largest magnification.

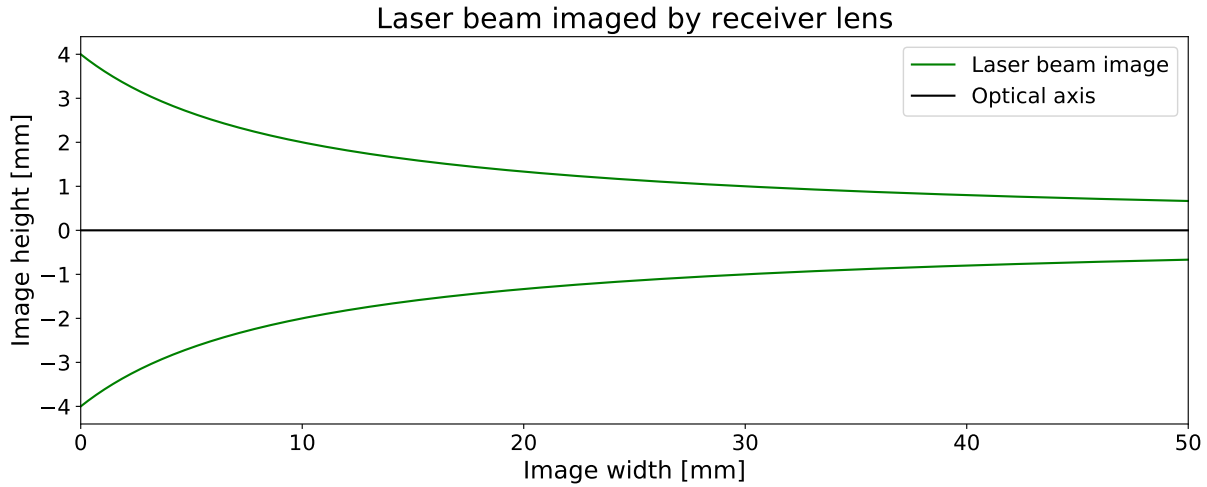


Figure 5.2: Shape of the image produced by the receiver lens using the inverse magnification relationship caused by the Scheimpflug configuration.

If such an image is seen on the detector when at the position of the slit, it will be easy to align the laser horizontally and with the optical axis with much greater precision that is quantifiable. Placing the slit at this location will thus confirm that a sharp image passes through the slit. The collimating lens can then be placed at a distance of f_{col} away from the slit along the optical axis. The PGP and filters can then be added, followed by the

focusing lens placed a distance of $f_{col} + f_{foc}$ away from the collimating lens. Using the same strategy as for aligning the slit, the detector can then record images to determine the location of the image plane after the focusing lens and sharply focus the slit.

The final finetuning would be to isolate the optical setup as much as possible. Nothing from the surroundings should hit the detector except what passes through the slit. This experiment used barriers to surround the optics and a tarp to cover the top. While it worked to some extent, it was not without flaws. An improvement can be made by creating dedicated boxes with openings only for connecting the optics. One box made for the transmitter, one box for the optics on the dovetail optical rail, and one for the detector. Creating such boxes will also make the experimental setup easier to transport. Boxes can be 3D-printed, built with LEGO, or a combination of the two.

5.2 Spectral analysis of soot

The candle image in Figure 4.1 confirmed that the setup worked and could be optimized for testing with soot. Spectral calibration image, such as Figure 4.4 demonstrated that the spectral interval covered the desired range. Figure 4.5 and Figure 4.8 showed that the soot generated of the mini-CAST could be detected. The spectra of OP1 soot and OP6 soot in Figure 4.6 and Figure 4.9 showed that the Raman spectra of soot can be spectrally resolved. Because of the nature of Eq.(2.15), the spectral resolution is not constant. At low values of p_λ , the spectral resolution is $\Delta\lambda = 5.7$ nm, while at larger values of p_λ it increases to $\Delta\lambda = 8.5$ nm, which differs from the spectral resolution of $\Delta\lambda = 4$ nm in the simulation given by Oskar. Yet, the spectral resolution was deemed sufficient to proceed with the project. These spectrally resolved Raman signals could be deconvolved, with varying certainty. Fourier analysis showed that the spectra could be split into polarized and unpolarized components, with the Raman signal from soot in the unpolarized spectra. These unpolarized spectra show clear removal of polarized components, however, with an increased signal-to-noise ratio for the DC spectra. The Fourier analysis should thus not be used to get a better spectrum, as a direct measurement with a polarizer gives the same result. This can be seen in Figure 5.3, where the DC signal from the Fourier analysis of OP1 soot is overlapped with the direct measurement during data collection, where the polarizer was perpendicular to the laser beam's polarization angle, blocking polarized components.

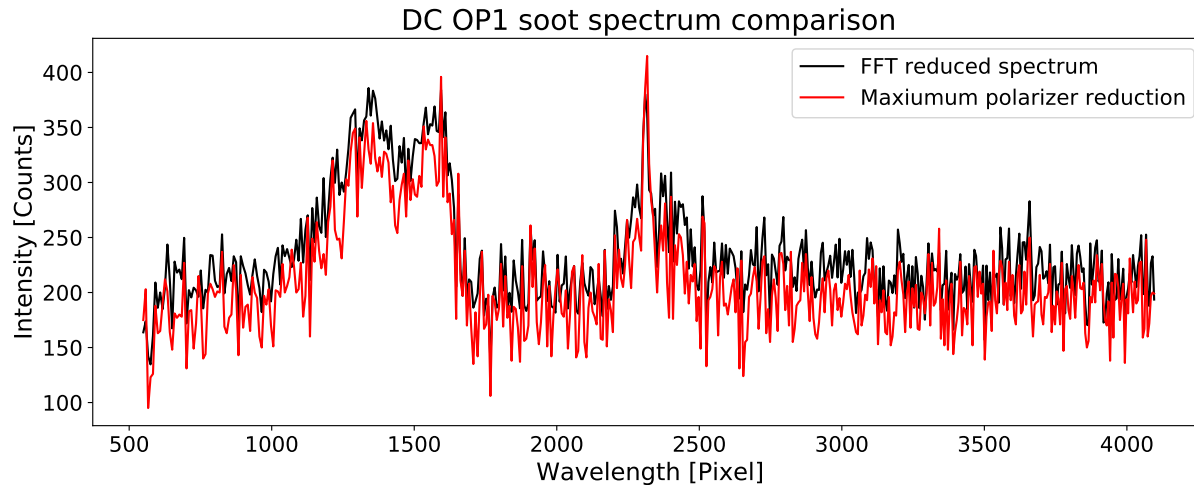


Figure 5.3: *Comparison between Fourier analysis and direct measurement with maximal polarized reduction from a polarizer, showing they are almost identical.*

As Figure 4.14 demonstrated, polarized gases can be identified using Fourier analysis and should be used for this purpose only. The reason the same separation was not successful for OP6 soot can be explained by a faulty experimental setup. Figure 5.4 shows one measurement from the Fourier analysis of OP6 soot. This shows a clear skewed soot signal, which was consistent across all measurements and was not realized until all measurements had been made. Since the data was inconsistent with what the Fourier analysis expects, it was not able to properly isolate the polarized components.

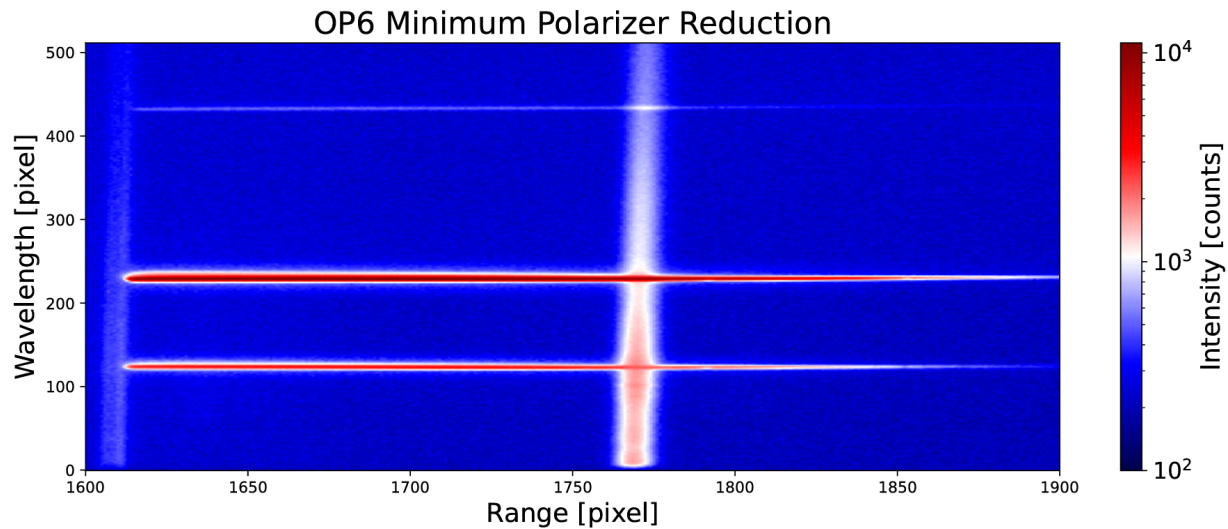


Figure 5.4: *The recorded image of skewed OP6 soot during Fourier analysis, where the polarizer is in a position that fully lets polarized light through.*

To test how well all fits are, both from direct and from Fourier analysis, a reduced χ^2 calculation can be used. Assuming that the intensity follows a Poisson distribution, the uncertainty of the intensity at a given frequency σ_ν is given by $\sigma_\nu = \sqrt{N_\nu}$ where N_ν is the intensity at frequency ν . The χ^2 is then determined by

$$\chi^2 = \sum_{\nu} \frac{(N_\nu - f_\nu)^2}{\sigma_\nu^2} \quad , \quad (5.2)$$

where N_ν is the observed intensity at ν and f_ν is the fitted value at ν . With multiple parameters fitted, it is better to determine the reduced χ^2 , which we denote by χ_r^2 and is defined as

$$\chi_r^2 = \frac{\chi^2}{m - p} \quad , \quad (5.3)$$

where m is the number of data points part of the spectrum, and p is the number of parameters used for the fitting. For each spectrum, the values of m , p , and χ_r^2 is provided in Table 5.1.

Table 5.1: χ_r^2 calculations for direct measurements of OP1 soot and OP6 soot, as well as for spectra on OP1 soot and OP6 soot after Fourier analysis removed polarized components.

Spectrum	m	p	χ_r^2
Direct OP1 soot	198	20	4.1
Direct OP6 soot	141	21	1.6
Fourier OP1 soot	198	11	2.2
Fourier OP6 soot	141	21	0.56

A value of $\chi_r^2 = 1$ is the target for a good model. As values stray away from it, either the model is incorrect, the data is imprecise, or the uncertainty estimation is wrong. Using the Poisson distribution for the DC spectra after Fourier analysis might lead to incorrect error estimation, since multiple spectra are involved in the calculation. A better approach to error estimation would be to assume a Poisson-distributed error before performing the Fourier analysis, rather than after, as was done in this project. Better data can be obtained by refining the experimental setup and reducing noise. A stronger laser will also increase the signal strength, resulting in more photons collected over the same time period.

6 Conclusion

This project experimentally investigated whether the hyperspectral Scheimpflug lidar could spectrally resolve the Raman signal from soot. The results from the experiment confirmed that it is possible by using both direct measurements and Fourier analysis with room for further improvements. The typical D and G peaks in Raman spectra of soot can be spectrally resolved, enabling further structural analysis. Fluorescence is strongly evident in brown soot, indicating the presence of organic compounds within its disordered structure. Not only soot, but also gases can be detected using our technique. Their strong polarization property helps to separate them from unpolarized soot signals using polarization effects combined with Fourier analysis. It also showed that the Fourier analysis of the unpolarized spectra of OP1 soot removed polarized components as much as a direct measurement using a polarizer set to minimize polarized transmission, hence the purpose of the Fourier analysis should be to identify gases in the air and combustion process. To conclude, a rough construction following the Scheimpflug lidar technique for ranging, combined with a PGP for spectral separation, proved effective for resolving the Raman spectra of soot despite imperfections in the experimental setup, suggesting that an improved setup will yield better results. Improvements would include more precise alignment of components, positions, and rotations, and the construction of isolating boxes to minimize scattering from the surroundings.

With an improved and more easily portable experimental construction, the lidar setup can be brought out to perform measurements *in-situ* in climate chambers for further improvements and testing of the construction with the hopes of further developing it to allow for online measurements on real-time sources such as combustion processes in factories, exhaust pipes, fires, and volcanic eruptions to investigate the gas contributions, type of soot, and combustion process. This will provide a better understanding of soot, which will help climate modeling and discussions on how to reduce pollution to improve air quality, animal habitats, and the global climate.

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Appendix A: Scheimpflug Lidar Construction

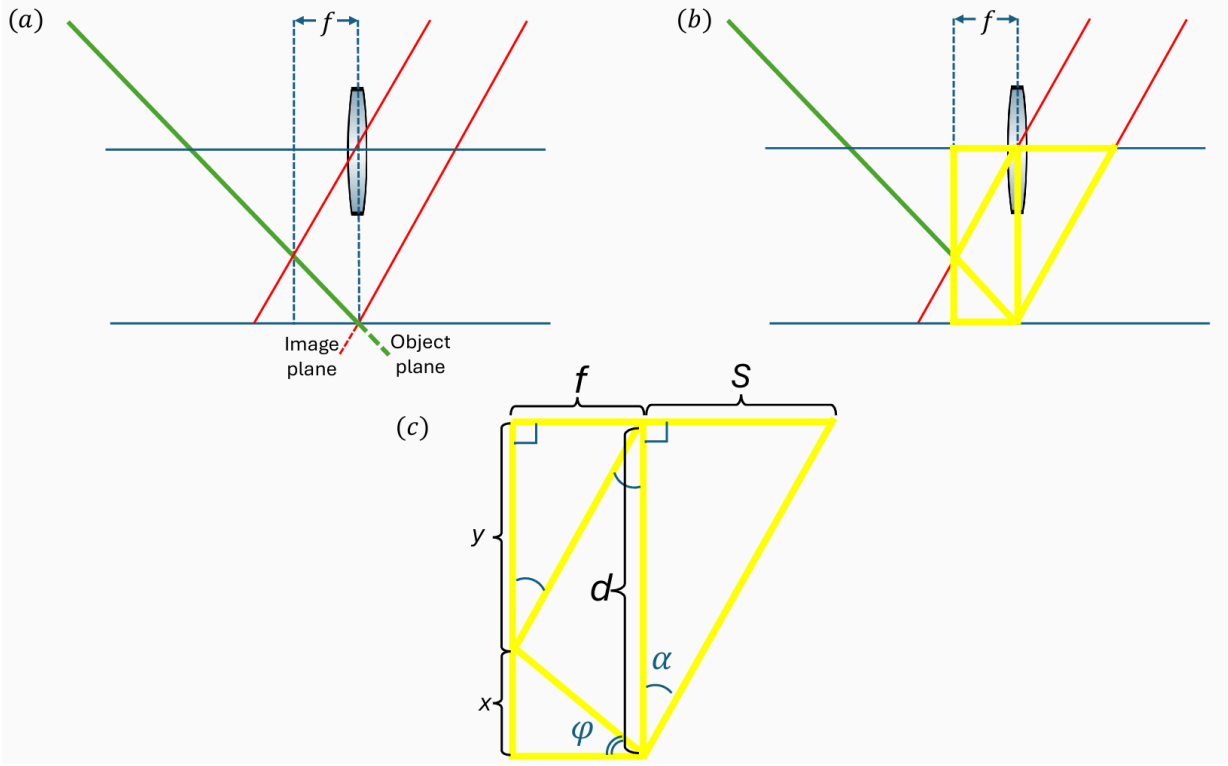


Figure A.1: (a) A figure that has placed out the object plane, image plane, and translated image plane relative to an imaging lens of focal length f . (b) The relevant geometry has been highlighted in yellow. (c) The relevant geometry has been isolated with sides and angles marked.

This derivation demonstrates how the image plane of a tilted object plane can be found using the Scheimpflug principle and the Hinge rule, without using the thin lens formula. In Figure A.1c, we have similar triangles that share two congruent angles, α and a right angle. From this, we can set up a similar triangle relation

$$\frac{S}{f} = \frac{d}{y} \quad . \quad (\text{A.1})$$

The side y can be expressed as $d - x$, meaning we get

$$\frac{S}{f} = \frac{d}{d - x} \quad . \quad (\text{A.2})$$

Since φ , f and x are part of a right triangle, they can be expressed together as

$$\begin{aligned} \tan(\varphi) &= \frac{x}{f} \\ \implies x &= f \tan(\varphi) \quad . \end{aligned} \quad (\text{A.3})$$

Substituting Eq.(A.3) into Eq.(A.2), we get

$$\begin{aligned} \frac{S}{f} &= \frac{d}{d - f \tan(\varphi)} \\ \implies S &= \frac{df}{d - f \tan(\varphi)} \quad , \end{aligned} \tag{A.4}$$

which concludes the derivation for Eq.(2.3). By dividing by d , we get the exact expression for $\tan(\alpha)$

$$\tan(\alpha) = \frac{S}{d} = \frac{f}{d - f \tan(\varphi)} \quad , \tag{A.5}$$

and thus

$$\alpha = \tan^{-1} \left(\frac{f}{d - f \tan(\varphi)} \right) \quad , \tag{A.6}$$

which concludes the derivation for Eq.(2.4).

Appendix B: SRL Ranging Formula

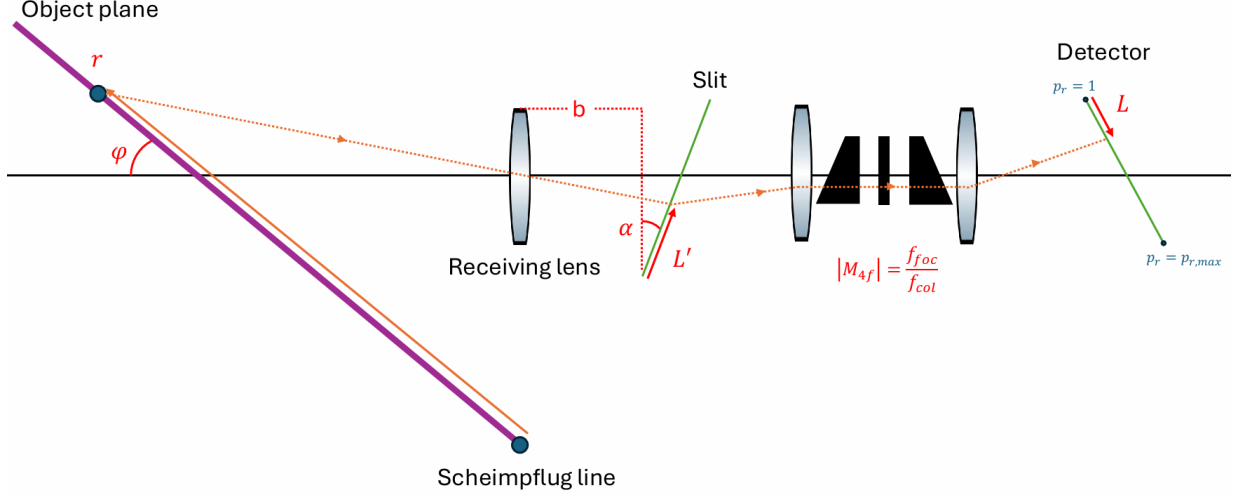


Figure B.1: *Illustration of how a point a distance r away from the Scheimpflug line gets imaged through a slit tilted at an angle α and past a 4f-optical system with magnification factor M_{4f} to be imaged onto pixel column p_r on a detector.*

For a given lens of known focal length f , a point a distance s_1 away from the lens on one side will be imaged a distance s_2 away from the lens on the other side. This can be done by using the thin lens formula

$$\frac{1}{s_1} + \frac{1}{s_2} = \frac{1}{f} \quad . \quad (\text{B.1})$$

For a tilted plane, such as the one presented in Figure B.1, the horizontal distance from a point along the plane to the lens plane is

$$s_1 = r \cos(\varphi) \quad , \quad (\text{B.2})$$

where r is the distance from the Scheimpflug line to the point, and φ is the slant angle of the object plane. Likewise, the distance s_2 can be derived from Figure B.1, which is

$$s_2 = b + L' \sin(\alpha) \quad . \quad (\text{B.3})$$

The distance b is defined in Figure 2.10 as the horizontal distance between the lens plane and the location of $p_r = 0$ on the slit. The tilt of the slit α is determined via the Hinge rule as defined in Appendix A. The distance L' is the distance along the slit between the points $p_r = 0$ and the point the object is mapped to. To convert the distance L' along the slit to the distance L along the detector, one simply needs to divide by the magnification factor of the 4f-optical system

$$L' \approx \frac{L}{|M_{4f}|} \quad , \quad (\text{B.4})$$

which becomes Eq.(2.8) if $L = l_{slit}$. The distance L along the detector will be the pixel size, l_{pix} , multiplied by the pixel column to the detected object, where the pixel column is defined in Figure 2.11

$$L = l_{pix} p_r \quad . \quad (B.5)$$

Substituting in Eq.(B.5) into Eq.(B.4) and then again substitute that into Eq.(B.3), the expression for s_2 will be

$$s_2 \approx b + \frac{l_{pix} p_r \sin(\alpha)}{|M_{4f}|} \quad . \quad (B.6)$$

Eq.(B.2) and Eq.(B.6) can then be used in the thin lens formula in Eq.(B.1)

$$\frac{1}{r \cos(\varphi)} + \frac{1}{b + \frac{l_{pix} p_r \sin(\alpha)}{|M_{4f}|}} \approx \frac{1}{f} \quad . \quad (B.7)$$

The goal is to obtain an expression for the distance r to a point as a function of the pixel column p_r on the detector to which it is mapped. This can now be done by solving for r in Eq.(B.7)

$$r(p_r) \approx \frac{f}{\cos(\varphi)} \left(1 + \frac{|M_{4f}| f}{|M_{4f}| (b - f) + p_r l_{pix} \sin(\alpha)} \right) \quad , \quad (B.8)$$

where p_r is determined on the detector, and b is calibrated, and all other parameters are known by the construction of the experiment. This concludes the derivation of Eq.(2.14).

Appendix C: Accepted ILRC Manuscript

Design and construction of a hyperspectral Scheimpflug Raman lidar for aerosolized soot ranging and characterization

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Abstract: In this study, we present a design and construction of a hyperspectral Scheimpflug Raman lidar for range-resolved characterization of aerosolized soot. For the first time in a laboratory-scale configuration, the system enables detection and discrimination of soot particles at different maturity levels at elevated distances. The optical setup provides high spectral resolution, allowing clear observation of the characteristic D and G Raman bands of soot. Optical ray-tracing optimization was performed using the software ZEMAX to determine the optimal placement of the system components, balancing spectral resolution with compact instrument design. A continuous-wave 532 nm laser with output power up to 2 W was used to detect Raman scattering from black and brown soot generated under different operating conditions using a mini-CAST soot generator. The results demonstrate the capability of hyperspectral Scheimpflug Raman lidar for remote, spectrally resolved soot characterization, highlighting its potential for controlled laboratory studies and future atmospheric applications.

1. Introduction

Although green energy is getting more popular and will be a long-term solution for our planet, non-green energy from various combustion processes of hydrocarbon such as fossil fuels, biomass is still the main energy source for mankind for many more years [1]. As a result of these incomplete combustion processes, carbonaceous substances, particularly soot (also known as black carbon), are produced and threaten human health and the earth's climate [2,3]. Quantitative and qualitative information of soot emission in the atmosphere plays an important role in predicting its negative effects so that timely solutions can be implemented.

By examining the Raman spectral properties of soot, insights can be obtained into its structural order and chemical composition [4–6]. The most prominent Raman features of soot are the so-called D (disorder or defect) and G (graphitic) bands, which are indicative of its structural organization [7]. The G band ($\sim 1580\text{ cm}^{-1}$) is associated with the in-plane vibrational modes of the graphitic carbon network, whereas the D band ($\sim 1360\text{ cm}^{-1}$) arises from structural defects and disorder within the graphitic lattice [8].

In the case of brown soot, which contains a higher fraction of toxic organic compounds and has been shown to exert more severe adverse health effects, strong fluorescence interference often overwhelms the Raman signal [5,6]. In contrast, black soot, characterized by a higher degree of graphitization and a lower organic content, exhibits weaker fluorescence but significantly stronger light absorption, leading to a greater contribution to climate change through radiative forcing [9].

In many real-world scenarios, soot sources are inaccessible for direct sampling or measurements at close distances, so conventional characterization and quantification approaches become impractical. Extending Raman-based soot analysis to remote sensing configurations is therefore essential for such emissions under realistic conditions. Traditional lidar relies on time-of-flight to do ranging using pulsed lasers. However, because Raman scattering is intrinsically weak compared to elastic scattering, detectable Raman signal typically requires high excitation power. In the case of soot, high peak laser power will lead to thermal distortion, structural modification or even destruction of the particles [6].

By employing the Scheimpflug optical configuration, the system can use a CW laser rather

than a pulsed laser [10], while still enabling ranging of soot and preserving its chemical structure.

2. Methodology

The Scheimpflug principle and the hinge rule are combined to achieve sharp focus over an extended tilted object plane. The Scheimpflug principle states that sharp focus across a tilted objective plane is obtained when the objective plane, lens plane, and image plane intersect along a common line (points S_1 , S_2 in Fig. 1) [11]. The hinge rule further specifies the position and orientation of the focused object plane. According to the hinge rule, when the image plane is translated to intersect the center of the imaging lens, this translated plane intersects the object plane at the same point where

it intersects with the front focal plane of the lens (point H_1 , H_2 in Figure 1) [12].

Fig. 1 shows the top and side view of the experimental setup, in which the Scheimpflug principle and the hinge rule are applied twice. In the first application, the tilted objective plane is imaged onto the entrance slit, which is positioned in the first image plane and serves as a secondary object plane. In the second application, the slit is imaged onto the detector. Here, the light emerging collimated from lens L_2 is spectrally dispersed using a prism-grating-prism (PGP) assembly before being focused by lens L_3 onto the tilted detector sensor.

In this work, the tilted object plane corresponds to the probe volume, defined by the laser beam path over the detected range, as illustrated in Fig. 1.

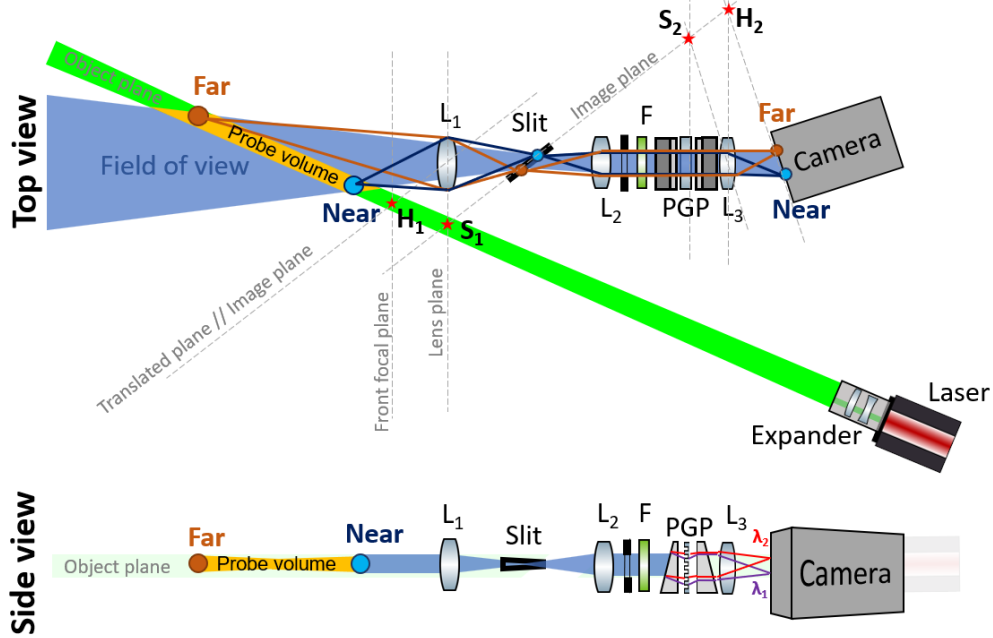


Fig. 1: Top and side views of the Scheimpflug Raman lidar. L : lenses, PGP: prism-grating-prism, F : notch filter and long pass filter for removing elastic.

Table 1 presents optical components and technical information of the experimental setup.

The combination of Scheimpflug lidar geometry with the PGP spectrometer enables simultaneous ranging and spectroscopic characterization of soot particles, with spatial range information encoded along the horizontal axis and spectral information resolved along the vertical axis of the detector.

For ranging calibration, we use the formula below [13]

$$r_{pix(n)} = d \frac{f_{rec} + n M l_{pix} (\sin \alpha - \cos \alpha \cot \varphi)}{f_{rec} \cot \varphi + n M l_{pix} (\sin \alpha \cot \varphi + \cos \alpha)} \quad (1)$$

where d is the baseline distance, f_{rec} is the receiver focal length, l_{pix} is the pixel size, and M is the slit-

to-detector magnification. The angles α and φ denote the slit tilt and the transmitter tilt, respectively. The index n ideally ranges from 1 to the total number of pixel columns. But due to the potential lateral offset in the detected image, the pixel index must be corrected to $n+n'$, where n' is a pixel that depends on the experimental alignment and setup. The shift n' can be determined by measuring the distance to a terminal target and the corresponding pixel column, and then fitting Eq. (1) to the measured distance-pixel relationship to retrieve n' . Spectral calibration is performed using well-known Raman fingerprints of ambient O_2 , N_2 and water vapor measured from the air.

Table 1. Technical information of the Scheimpflug Raman lidar

Component	Technical information
Transmitter	
Laser	532 nm, 2 W, CW
Slant angle	5.5°
Receiver	
Receiving lens (L ₁)	f = 200 mm
Slit (length × width)	50 × 0.2 mm
Slit tilt	~ 46.8°
Collimating lens (L ₂)	f = 150 mm
Filters (F)	Notch filter (NF533-17) & Long pass filter (FELH0550)
Prism	600 grooves/mm
Grating	18.15° (wedge angle)
Focusing lens (L ₃)	f = 80 mm
Slit-to-detector magnification	~ 2:1
Detector	Andor CCD DU940NBV (2048 × 512 pixel, 27.6 × 6.9 mm ²) pixel size 13.5 × 13.5 μm ²
Detector tilt	33.3°
System	
Baseline separation	200 mm
Range	1 – 10 m
Spectral resolution	4 nm
Spectral range	540 – 642 nm

3. Result and discussion

With the primary setup parameters defined, ray-tracing simulations were performed in ZEMAX. Fixed parameters are listed in Table 1, while the receiver optics distances were optimized to minimize the detector spot size, ensuring sufficient spectral resolution to resolve the D and G bands of soot. The optimized configuration was then implemented experimentally with minor adjustments to accommodate practical alignment constraints, with particular focus on optimizing the entrance slit and detector positions.

Figure 2 illustrates the operation of the receiver optics as modeled in ZEMAX, with component parameter imported directly from Thorlabs libraries. As shown, the detected light is spectrally dispersed vertically, as indicated by the different colors, while spatial separation occurs horizontally due to the Scheimpflug geometry. To maximize optical throughput, the distances between the grating and the two prisms in the PGP were positioned as closely as possible. The distances between other components between the collimating and focusing lens, i.e. the iris and filters, did not require further optimization since the light is collimated in this region and these components do not significantly affect spectral resolution.

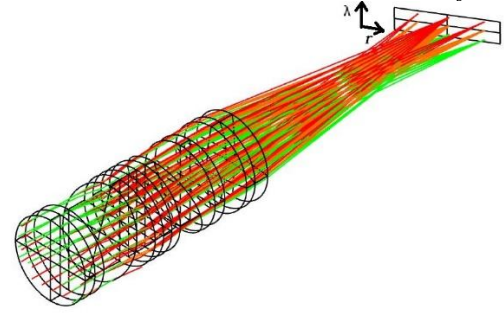


Fig. 2: ZEMAX simulation of the optimized receiver optics. Different colors represent different wavelengths. Spectral dispersion and ranging are observed in the vertical and horizontal directions on the detector, respectively.

The vertical separation on the detector between spot images corresponding to two wavelengths differing by 4 nm is shown in Fig. 3. This result demonstrates that the achieved spectral resolution is sufficient to resolve the D and G bands of soot, given that the detector pixel size is 13.5 × 13.5 μm², and that the wavelength separation between the D and G bands are approximately 8 nm.

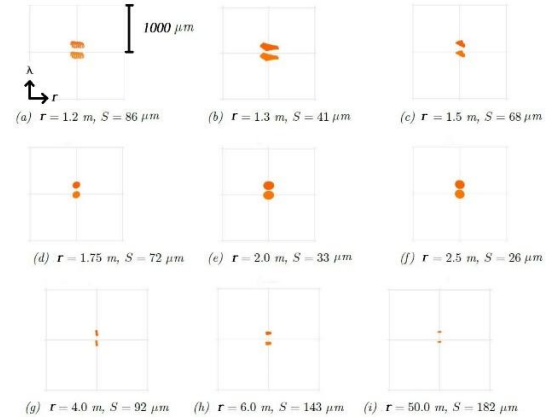


Fig. 3: ZEMAX simulation of spot sizes at various range (r). The wavelengths used here are 591 nm (lower spot) and 595 nm (upper spot). S is the vertical separation between the spots.

It is noteworthy that the spectral resolution varies with range. While the spectral resolution is higher at both short ranges (e.g., Fig. 3d) and longer ranges (e.g., Fig. 3h), the largest spot sizes (and thus worst spectral resolution) occur at mid-range around 2 m. That could be a consequence of aberration because different ranges map to different field points in the Scheimpflug imaging geometry, leading to range-dependent defocus and aberrations.

The construction of the experimental setup followed the predetermined parameters of the optical components, as well as the optimized distances between them obtained from the ZEMAX simulations. Fig. 4 shows range-resolved Raman spectrum of soot generated by the mini-CAST soot generator, recorded in a dark room

environment with an exposure time of 50 s and 40 accumulations. The laser was horizontally polarized, and no polarizer was included in the detection path.

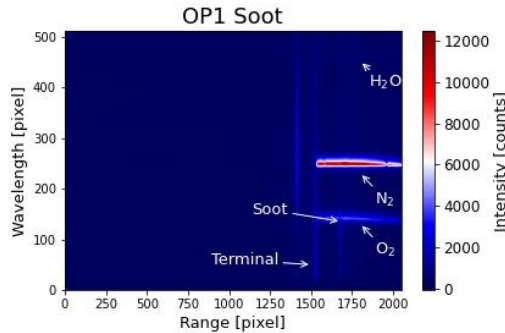


Fig. 4: Raman spectra of OP1 soot and ambient gases over the range up to 10 m in the laboratory.

Spectroscopic measurements of soot in the atmosphere are affected by Raman scattering from air, primarily due to N_2 and O_2 and H_2O along the detection range. In particular, the O_2 Raman band at 1556 cm^{-1} interferes with the soot G band at approximately 1580 cm^{-1} , as illustrated in Fig. 5. In addition to Raman signals, fluorescence background is observed and is significantly stronger for OP6 soot (brown soot) than for OP1 soot (black soot). This enhanced fluorescence contribution provides a spectral signature that can be used to distinguish between different soot types.

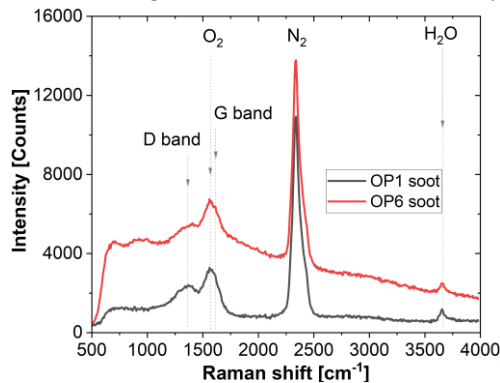


Fig. 5: Raman spectra of OP1 soot (replotted from Fig. 4) and OP6 soot at the range of 2 m.

The contribution of N_2 , O_2 and H_2O can be reduced if the polarization of the laser beam and the collected scattered photons are perpendicular. It should then be possible to de-convolve the D and G peaks respectively to gain insight to the structure of the detected soot. Our recent study shows that the vibrational Raman modes of O_2 and N_2 are strongly polarized while Raman spectra of soot are nearly unpolarised [14,15]. Therefore, the spectral contributions of the air could be suppressed using polarization analysis.

4. Conclusion and outlook

This work demonstrates the feasibility of range-resolved characterization of aerosolized soot using

a Scheimpflug Raman lidar. By adjusting the slant angle as well as the slit and detector tilts, the accessible measurement range can be extended to further distances while maintaining sufficient spectral resolution. Polarization-resolved detection can be implemented to suppress interference from molecular Raman scattering, particularly the oxygen Raman band that overlaps with key soot Raman features.

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