DEVELOPMENT OF A SPATIALLY RESOLVED OPTICAL TECHNIQUE TO MEASURE TEMPERATURE USING TWO-PHOTON ABSORPTION OF XENON

By

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ABSTRACT

Development of a Spatially Resolved Optical Technique to Measure Temperature using Two-Photon Absorption of Xenon

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A new spatially resolved optical technique to measure temperature was developed using two-photon absorption of xenon. This experiment excited the 256 nm two-photon transition of xenon by focusing the excitation source into a test cell at room temperature. Two-photon absorption only occurs at the focus of a laser beam; therefore a point measurement technique was possible. The spatial resolution for this experiment was approximately 0.6 mm; however, this technique enables higher resolution depending on the focus of the laser beam. Two-photon absorbance versus xenon number density was determined experimentally and used to validate a theoretical model created in MATLAB. This technique was designed primarily for two non-reacting flows: the vortex tube and the pulse tube. A case study for measurement in a vortex tube was presented; however, applying this technique to a pulse tube will be similar.
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1. INTRODUCTION

1.1 Motivation

The vortex tube and the pulse tube are both simple devices that operate with non-reacting flow. The vortex tube is a cooling device that uses no moving parts and separates an inlet flow into two streams at different temperatures [1]. The vortex tube has been recently developed commercially for spot cooling applications [2], such as cooling electronic controls or cooling machining operations. The pulse tube system is classified as a small cryogenic refrigerator [3], and the advantage of the pulse tube, similar to the vortex tube, is the simplicity of design since the pulse tube has no moving parts. The pulse tube provides a way for the oscillatory gas flow to carry the heat from the cold source to the hot source, i.e. the refrigeration of the system [4]. For both of these devices the thermal-fluid processes are not completely understood due to the lack of information regarding the gas flow properties, and each device would benefit from a method to obtain point measurements of temperature. Therefore, a spatially resolved optical technique for measuring temperature was developed primarily for the vortex tube and the pulse tube.

Thermocouples and other similar devices are the conventional method to measure the temperature of a gas, liquid or solid. However, for the vortex tube and pulse tube, the heat transfer is driven by the interactions of the fluid, and the presence of a thermocouple would disturb the fluid flow and create changes in the temperature distribution. This is also the case for reacting flows, for example in a research flame; a thermocouple changes the flame properties causing inaccurate temperature measurements [5]. To circumvent
these limitations, “optical thermocouples” are being developed which offer improved performance over the conventional probe since they are completely non-intrusive, meaning they do not change the environment under investigation. Along with the advantage of non-intrusiveness, several optical techniques have been demonstrated to make point measurements of temperature [6]. Accordingly, optical techniques are providing point measurements of temperature in systems that were not previously possible.

1.2 Existing Techniques

Raman scattering, Rayleigh scattering, laser-induced fluorescence (LIF), and coherent anti-stokes Raman scattering are the main optical techniques providing point measurements of temperature. Raman and Rayleigh scattering are both non-resonant techniques, meaning they can be performed using a laser of arbitrary wavelength. Under ideal circumstances, Rayleigh scattering is particularly attractive, because measurement of a single intensity can provide gas density, and temperature can then be inferred by use of an equation of state for the gas. Recently, the Rayleigh scattering technique has been used to measure temperature of fluid within a pulse tube [7]. However, because Rayleigh and Raman scattering are non-resonant, the signal is weak and scattering from surfaces or particles often interfere with the desired signal. Furthermore, in reacting flows, the signal can become indecipherable because Rayleigh scattering depends strongly on gas composition. Resonant techniques such as laser-induced fluorescence (LIF) provide a higher signal and target only a single species. However, the species most amenable to LIF, for example, OH, NO, formaldehyde, and acetone [8] may not be present naturally.
Seeding of these species may be undesirable; for example, NO is toxic and acetone’s vapor pressure may be too low for flows below ambient temperature since there is interest in operation of pulse tubes at cryogenic temperatures. Even when an appropriate tracer is present, temperature measurements by LIF may require multiple lasers. Nonlinear techniques such as coherent anti-stokes Raman scattering (CARS) provide a strong signal in the form of a laser beam, and allow access to inert gases such as nitrogen [9]. The two-photon absorption technique presented here shares these advantages with CARS, but is relatively simple. However, unlike CARS, the technique is resonant and thus generally requires a wavelength-tunable laser.

1.3 Two-Photon Absorption of Xenon

This paper demonstrates a new spatially resolved optical technique to measure temperature: two-photon absorption of xenon. Compared to the non-resonant techniques, two-photon absorption of xenon is advantageous because the signal is much stronger and there is a visual indicator of the point measurement location as a result of laser induced fluorescence. While this technique is primarily developed for two non-reacting flows, two-photon absorption of xenon could be applied to many different systems, including reacting flows. Xenon is a noble and inert gas; therefore, it has extremely weak tendencies to chemically react with other materials. This allows xenon to be used as a tracer in reacting flows without altering the chemical processes.

There are a few disadvantages of two-photon absorption of xenon compared to the other spatially resolved optical techniques. First, xenon is an expensive gas mostly limiting
this technique to closed environments or environments in which the xenon can be recovered and reused. There is potential to develop this technique for other inert gases, such as argon; however, xenon will be used initially since it has a strong two-photon absorption signal. Second, two-photon absorption is somewhat sensitive to window fouling as described in Sections 7.1 and 7.2 and therefore performs best in clean environments. Two-photon absorption is a non-linear process and different results will occur depending on whether the transmitted irradiance decreases before or after the point measurement. Third, two-photon absorption is sensitive to beam steering; beam steering will generally result in reduced absorption, which could incorrectly be interpreted as reduced xenon density.

Despite the disadvantages, this experiment demonstrated the two-photon absorption technique by exciting the 256 nm two-photon transition of xenon inside a test cell at room temperature. The excitation source was focused inside the test cell since two-photon absorption only occurs at the focus of a laser beam. The spatial resolution of this technique was determined by the focusing parameters of the laser beam. For this experiment at a focusing angle of 0.96 deg, the spatial resolution was approximately equal to 0.6 mm. Since the pressure and temperature was known for this experiment, two-photon absorbance as a function of xenon number density was determined. Using these results, a point measurement of temperature as a function of time for a device of interest can be attained by making continuous measurements of xenon number density and inferring temperature from the ideal gas law for a known pressure.
1.4 Thesis Objective

The overall goals of this project were to develop a spatially resolved technique to measure temperature by two-photon absorption of xenon, to validate the theoretical model, and to apply this technique to either a vortex tube or pulse tube. This paper mostly focuses on the first two goals. While this technique was never directly applied to the vortex tube or pulse tube, this paper proposes an optical setup and a method for making continuous point temperature measurements in a vortex tube.
2. BACKGROUND

2.1 Two-photon absorption

An atom or molecule has certain, discrete energetic states. To transition from one state to another, an atom or molecule can gain a specific amount of energy by absorbing a photon of light at a particular wavelength. To reach the same excited state a species can simultaneously absorb two photons of light of half the energy and twice the wavelength. Two-photon absorption can be pictured by excitation from an initial state to a virtual state and then to a final state as shown in Figure 1.

Two-photon absorption is considered one of the simplest non-linear techniques. The first to develop the theory of two-photon absorption was Maria Göppert-Mayer in 1931 in her Ph.D Thesis (see [10] and references therein). The probability of a two-photon transition to occur is proportional to the light irradiance; therefore, an intense light source is needed. As a result, the first observation of a multi-photon process did not occur until 1961 (see [11] and reference therein). Two-photon absorption only occurs at the focus of a laser beam where the irradiance is high; therefore, a point measurement technique is possible by focusing a laser beam at a certain wavelength to excite a multi-photon transition for an atom or molecule.

Xenon has two-photon transitions at 249.63 nm, 256.02 nm, and 252.49 nm [12]. This experiment excites the $5p^6 1S_0 \rightarrow 5p^5 6p [5/2]_2$ xenon absorption line with two photons of 256 nm. The excitation wavelength of 256 nm is selected based on the strength of the
two-photon transition. To reach the same excited state, one photon of 128 nm light is required; however, excitation with 128 nm light is not possible since air absorbs this wavelength. Thus, there are two fundamental advantages of the two-photon approach: 1) it allows access to transitions that would otherwise be possible only in the vacuum ultraviolet and 2) it allows a point measurement by appropriate focusing.

![Figure 1. One-photon and two-photon excitation to a higher energy level of xenon.](image)

### 2.2 Spatial Resolution

The spatial resolution for two-photon absorption is defined by Gaussian parameters. By focusing a Gaussian laser beam to the diffraction-limited spot size, the spatial resolution is approximately equal to two times the Rayleigh range shown in Figure 2 as the absorption length, \( l \). Figure 2 is modified from a diagram in reference [13].

![Figure 2. Absorption length, \( l \), defined by Gaussian parameters.](image)
The Rayleigh range is defined in reference [13] and shown in Equation 1.

\[ Z_R = \frac{\pi \omega_0^2}{\lambda} \]  \hspace{1cm} (1)

Where \( \lambda \) is the laser beam wavelength and \( \omega_0 \) is the beam waist (or beam radius) defined at \( 1/e^2 \) for intensity. The beam waist is located at the focus point and represents the smallest theoretical spot size.

As a result, the spatial resolution of this technique is limited by the laser beam and the focusing optics. For a Gaussian laser beam, 2 mm in diameter and focused by a 20 mm focal length lens, the minimum beam waist at the focus is 1.5 \( \mu m \) and the Rayleigh range is equal to 27 \( \mu m \). Thus, two-photon absorption will only occur over a total distance of \( \sim 54 \mu m \), making this a fairly high resolution technique.

By varying the focusing angle with the laser beam diameter or the focal length of the lens, the spatial resolution can be selected to meet the point measurement goals. For instance, by focusing a 2 mm diameter laser beam with a longer focal length lens of 100 mm, the absorption length increases to 1.4 mm.
3. EXPERIMENTAL DESIGN

3.1 Optical Setup

Figure 3. Optical setup for measuring two-photon absorbance of xenon at a point location inside a test cell.

Figure 3 shows a schematic diagram of the experimental setup for measuring the amount of two-photon absorption in xenon. The excitation source was a titanium:sapphire oscillator (Spectra Physics Tsunami, pumped by Millennia X) emitting short duration pulses at 768 nm. The estimated pulse duration was 54 fs (calculation for the pulse duration is shown at the end of this section), and the pulses were delivered at a repetition rate of 80 MHz. The laser average power was approximately 1.3 W and the spectral full width at half maximum was 15 nm. The Ti:sapphire output was first sent through a
neutral density filter before the beam was sent through a third-harmonic generation (THG) package (U-Oplaz Technologies, TP-2000B fs tripler). The filter was used to prevent damage of the THG optics. The tripler produced frequency doubled, $2\omega$, and tripled, $3\omega$, collimated light at 384 nm and 256 nm, respectively, each with approximately 50 mW average power and 120 fs pulse duration. The $2\omega$ beam was blocked and not used in this experiment. The $3\omega$ beam power was first recorded by placing a power meter (Melles Griot 13PEM001/J 2-Watt Broadband Power/Energy Meter) in the beam path. After the power was recorded the $3\omega$ beam entered the test cell, which was a 50.8 mm inner diameter Swagelok union tee fitted with UV fused silica windows. The test cell was pressurized with xenon and nitrogen gas, and the total pressure in the test cell was recorded by a pressure gauge connecting the gas tanks. Inside the test cell, a plano-convex lens (Thorlabs LA4725), L1, with a 75 mm focal length was used to focus the beam. At the focus of L1, the $3\omega$ beam was absorbed by the xenon gas and refocused by lens, L2, onto a power meter. Recording the power before and after the test cell allowed measurement of the initial irradiance, $I_0$, and the irradiance over the absorption length, $I_{l}$, located before and after the focus of L1.

To verify two-photon absorption of xenon was occurring at the focus, L1 was positioned inside the test cell, allowing the focus point to be visible by the window perpendicular to the beam path. By looking into this test cell window a green fluorescence spot was visible at the focus location of L1 during the two-photon absorption measurements. To record the fluorescence from the focus of L1, the light was collected by a bi-convex lens, L3, and focused into a 550 µm fiber. The fluorescence spectrum was recorded on a
spectrograph (Thermo Oriel, MS260i ¼ m dual grating spectrograph) and the results are given in Section 10.

The pulse duration of Spectra Physics Tsunami Ti:sapphire laser was estimated based on the transform-limit for a hyperbolic secant pulse shape model [14] as shown in Equation 2.

\[ \Delta t_p = \frac{0.315}{\Delta \nu} \]  

(2)

Where \( \Delta t_p \) = transform-limit pulse duration [s]

\( \Delta \nu \) = spectral full width at half maximum of the pulse [Hz]

The spectral full width at half maximum (FWHM) of the pulse, \( \Delta \nu \), was equal to 7.6 THz for a FWHM of 15 nm at a wavelength of 768 nm. Thus, the Ti:sapphire laser transform-limit pulse duration was equal to 41 fs. To estimate the actual pulse duration, the manufacturer data recommends multiplying the transform-limit pulse duration by the factor 1.3; therefore, the actual pulse duration emitted from the Spectra Physics Tsunami Ti:sapphire laser was approximately 54 fs as stated above.
4. THEORY

4.1 Development of Theoretical Model

An equation presented elsewhere [15] was used to calculate the irradiance of the laser beam over the absorption length, \( l \), as shown in Figure 2, Section 2.2. The derivation of this equation is based on the assumptions of a top hat radial and temporal profile.

\[
I_i = \frac{I_0}{1 + l/l_0} \tag{3}
\]

The absorption length, \( l \), or distance over which the two-photon absorption occurs is approximated by twice the Rayleigh range given in Equation 1, Section 2.2. Therefore, the absorption length is defined by Gaussian beam parameters shown in Equation 4.

\[
l = \frac{2\pi \omega_0^2}{\lambda} \tag{4}
\]

Where, \( \omega_0 \) = Diffraction-limited beam waist for a Gaussian beam defined at 1/e\(^2\) for intensity [m]

\( \lambda \) = Laser wavelength [m]

The laser for this experiment was the frequency tripled, 3\( \omega \), light from the tripler unit at a wavelength, \( \lambda \), of 256 nm (shown in Figure 3). The 3\( \omega \) beam had an elliptical spot profile (y-direction diameter \( \sim 2.3 \) mm and x-direction diameter \( \sim 6 \) mm); however, this paper assumed the 3\( \omega \) beam was circular with a diameter of 2.3 mm. The smaller beam diameter was assumed because it causes the largest diffraction-limited beam waist, \( \omega_0 \), equal to 4.8 \( \mu \)m. The diffraction-limited beam waist, \( \omega_0 \), was estimated by using a ray tracing program, ZEMAX Optical Design Program (see Appendix III a, Equation 22).
The length at which the irradiance has decreased to 50% of its initial value is given by Equation 5.

\[ I_0 = \frac{hc^2\nu_1}{2\sigma GCl_0N_1} \quad (5) \]

Where,

- \( I_0 \) = Initial laser irradiance [W/m\(^2\)]
- \( h \) = Planck’s constant \( \approx 6.63 \times 10^{-34} \) [J•s]
- \( c \) = Speed of light in a vacuum \( \approx 3 \times 10^8 \) [m/s]
- \( \nu_1 \) = Laser wavenumber [m\(^{-1}\)]
- \( \sigma \) = Two-photon absorption cross-section [m\(^4\)]
- \( G \) = Second-order coherence function of the photon field
- \( C \) = Convolution of the laser line profile [m]
- \( N_1 \) = Number density of ground-state atoms [atoms/m\(^3\)]

For a pulsed laser, the initial laser irradiance is defined by Equation 6.

\[ I_0 = \frac{P_0}{A_c f \Delta t} \quad (6) \]

Where,

- \( P_0 \) = Initial average laser power [W]
- \( A_c \) = Cross-sectional area at beam waist [m\(^2\)]
- \( f \) = Laser pulse repetition rate [Hz]
- \( \Delta t \) = Laser pulse duration [s]

Equation 7 states that the cross-sectional area is a function of the factor \( M^2 \), which will be defined and calculated in Section 4.2 and 4.3, respectively. This factor relates the actual beam waist to the diffraction limited beam waist, \( \omega_0 \).

\[ A_c = \pi M^2 \omega_0^2 \quad (7) \]

The convolution of the laser line profile is defined by Equation 8.
\[ C = \sqrt{\frac{4\ln(2)}{\pi \left(2(\Delta \nu_1)^2 + (\Delta \nu_{13})^2\right)}} \]  

(8)

Where,  
\[ \Delta \nu_1 = \text{full width at half maximum of laser line spectrum} \ [m^{-1}] \]
\[ \Delta \nu_{13} = \text{full width at half maximum of the absorption spectrum} \ [m^{-1}] \]

The full width at half maximum of the absorption spectrum, \( \Delta \nu_1 \), was assumed negligible since it is extremely small compared to the laser line spectrum. The full width at half maximum of the laser spectrum equaled 15258.8 m\(^{-1}\) (see Appendix IV).

Finally, the amount of two-photon absorbance of xenon is derived from Beer-Lambert law [16] given by Equation 9 below.

\[ \alpha_{\text{Xe}} = -\ln\left(\frac{I}{I_0}\right) \]  

(9)

The amount of two-photon absorbance of xenon as a function of xenon number density was plotted in Figure 4 for the following input parameters: an average input power, \( P_0 = 100 \text{ mW} \), xenon mole fraction, \( x = 1 \), a laser pulse duration, \( \Delta t_p = 308 \text{ fs} \), \( M^2 \) value of 1, second-order coherence function of the photon field, \( G = 1 \) [17], an absorption cross-section, \( \sigma = 4.0 \times 10^{-43} \text{ m}^4 \) [12], and a convolution of the laser line profile assuming Gaussian profile, \( C = 4.353 \times 10^{-5} \text{ m} \). For a temperature of 298 K, this plot corresponded to a pressure range of 0 to 689 kPa. Therefore, at room temperature and 689 kPa, the theoretical code predicted 58% two-photon absorbance of xenon. Note, there is a non-linear dependence of the absorbance on xenon number density.
Figure 4. Theoretical two-photon absorbance as a function of xenon number density for a particular set of input code parameters.

Figure 4 gives an estimate for the amount of two-photon absorbance of xenon expected in the test cell for a constant initial irradiance. For experimental measurements, the laser may vary and in order to compare the experimental and theoretical results, a new parameter is derived from Equations 3 and 5, and this new parameter will be referred to as beta ($\beta$).

$$\beta = \left(1 - \frac{1}{I_i} \right) = \frac{2l\sigma G C N_i}{hc^2 v_i}$$

(10)

Accordingly, beta has units of m$^2$/W. From the theoretical code, beta as a function of xenon number density was plotted in Figure 5. This theoretical result was based on the following code parameters: xenon mole fraction, $x = 1$, two-photon absorption cross-section, $\sigma = 4.0 \times 10^{-43}$ m$^4$, second-order coherence function of the photon field, $G = 1$, convolution of the laser line profile assuming Gaussian, $C = 4.353 \times 10^{-5}$ m, and an
absorption length, $l$, of 0.565mm. The plot shows that beta is independent of initial irradiance and $M^2$, and beta is a linear function of xenon number density; therefore, beta is a more suitable parameter for experimental measurements. For a temperature of 298 K, this plot corresponds to a pressure range of 0 to 689 kPa.

![Theoretical Beta vs Xenon Number Density](image)

**Figure 5.** Theoretical beta, $\beta$, versus xenon number density for a particular set of input code parameters.

The theoretical result shown in Figure 5 is valid for all temperatures. To determine a point measurement of temperature, the initial laser irradiance, $I_0$, and the irradiance of the laser beam over the absorption length, $I_l$, will be determined experimentally to calculate beta and xenon number density was inferred from Figure 5. With concentration and pressure known, temperature was calculated from the ideal gas law as shown below.

$$T = \frac{X_{Xe}P}{kN_{Xe}}$$  \hspace{1cm} (11)
Where, $X_{Xe} = \text{Mole fraction of xenon}$

$$P = \text{Total pressure [Pa]}$$

$$k = \text{Boltzmann constant} \sim 1.38 \times 10^{-23} \text{[J/K]}$$

$$N_{Xe} = \text{Xenon number density [atoms/m}^3\text{]}$$

### 4.2 Real Laser Beams

A real laser beam has both a waist and divergence angle which are larger than the ideal Gaussian parameters. The factor, $M^2$, was used to define the relationship between a real beam and an ideal Gaussian beam ($M^2 = 1$). The following equations derived in reference [13] show this relationship based on the divergence angle and beam waist.

$$\omega_{\text{real}} = M \omega_{0} \quad (12 \text{a})$$

$$\theta_{\text{real}} = M \theta \quad (12 \text{b})$$

$$M^2 = \frac{\omega_{\text{real}} \theta_{\text{real}} \pi}{4 \lambda} \quad (12 \text{c})$$

Where, $M = \text{Factor relating the real beam and ideal Gaussian beam}$

$$\omega_{\text{real}} = \text{Real beam waist (or beam radius) defined at } 1/e^2 \text{ for intensity}$$

$$\omega_{0} = \text{Gaussian beam waist (or beam radius) defined at } 1/e^2 \text{ for intensity} (M = 1)$$

$$\theta_{\text{real}} = \text{Real half angle of divergence defined at } 1/e^2 \text{ for intensity}$$

$$\theta = \text{Gaussian half angle of divergence defined at } 1/e^2 \text{ for intensity} (M = 1)$$

Two-photon absorbance is dependent on the factor $M^2$ as shown in Figure 6. This plot was for the same code input parameters given for Figure 4 in Section 4.1; however, the $M^2$ value was varied. As expected, the highest amount of two-photon absorbance for the system will occur for an ideal $M^2$ factor of 1.
4.3 Measurement of $M^2$

The output of the Ti:sapphire laser was estimated to have a $M^2$ value <1.2 based on manufacturer’s data. In the lab, the radial distribution of the Ti:sapphire laser was measured and compared to a Gaussian fit (see Appendix V, Figure 30 and Figure 31). While the $M^2$ value can not be directly determined from the Gaussian fit, the Ti:sapphire laser radial distribution showed a close agreement to a Gaussian curve indicating, similar to the manufacturer data, a small $M^2$ value.

However, as the Ti:sapphire laser was sent through the tripler unit and focused by the lens, L1, inside the test cell (see Figure 3), the $M^2$ value increased. To estimate the $M^2$
The collimated $3\omega$ beam was focused with a plano-convex lens, Thorlabs LA4725. This was the same lens as the optical setup in Figure 3. It is important to note that spherical aberrations exist due to the lens, and Section 5.2 shows the $M^2$ based on spherical aberration is equal to 2.677. At the focus of the lens a 10 µm pinhole was positioned on a translation stage and optimized to the focus location. The pinhole was selected to closely match the calculated diffraction-limited beam waist, $\omega_0$, of 4.8 µm, which was the largest beam waist of the $3\omega$ beam (see Appendix III a). Next, the average power before the pinhole, $P_1$, and the average power after the pinhole, $P_2$, was recorded with the Melles Griot power meter. The average powers, $P_1$ and $P_2$, equaled 18.4 mW and 4.2 mW, respectively.

Based on calculations performed with ZEMAX Optical Design, the ratio of $P_2/P_1$ was found to equal 0.837 for an $M^2$ factor equal to 1. Therefore, the $M^2$ value for the $3\omega$ was estimated to be 3.639. This $M^2$ value will be used to evaluate the measured two-photon
absorbance and beta, as well as the theoretical two-photon absorbance. Note that the theoretical beta is not dependent on $M^2$.

This approach for measuring the $M^2$ value only gave a rough estimate of this factor, $M^2$. There was uncertainty of this measurement based on the pinhole matching the diffraction-limited beam waist. The $3\omega$ beam had an elliptical spot profile; therefore, the largest diffraction-limited beam diameter was estimated to be 9.6 µm. A 10 µm pinhole was the smallest size available and used for this experiment.
5. EXPERIMENTAL CHALLENGES IN THE UV

5.1 Chromatic Dispersion

One of the challenges working with ultrafast UV light is chromatic dispersion. The index of refraction for a material varies with wavelength, thus the travel time through a material will depend on wavelength as well [18]. This is a problem working with short pulse durations, because the laser pulse will be stretched in time as it propagates through various optics. To quantify the amount of chromatic dispersion, a coefficient is measured in units of picosecond (of laser pulse duration) per nanometer (of wavelength bandwidth) per kilometer (of optical material).

The lenses, L1 and L2, in the optical setup (Figure 3) were UV fused silica material which features high internal transmission in the UV (at 256nm ~99.8%). For UV fused silica the dispersion coefficient is -6151 ps/nm/km for 256 nm, -1320.5 ps/nm/km for 384 nm, and -122.53 ps/nm/km for 770 nm [19]. For the purpose of comparing chromatic dispersion over a broad wavelength range, the following bandwidths (FWHM) at the certain wavelength were chosen to compare: 1 nm at 256 nm, 1.5 nm at 384 nm, and 3 nm at 770 nm, which were selected on the basis of bandwidth to wavelength ratio equal to 0.39%. This ratio was selected because the frequency tripled, $3\omega$, beam at 256 nm had a spectral full width at half maximum equal to approximately 1nm (see Appendix IV).

Figure 8 shows a plot of pulse duration as a function of UV fused silica material thickness for the selected wavelengths: 256 nm, 384 nm, and 770 nm. The pulse
duration of the $3\omega$ beam was approximately 120 fs, shown as the initial pulse duration for Figure 8. Due to chromatic dispersion, the pulse duration increased as the laser beam passes through more fused silica material width and the effect was dramatic for UV wavelengths (256 nm and 384 nm).

![Chromatic Dispersion for Initial Pulse Duration of 120fs](image)

**Figure 8.** Pulse duration versus UV fused silica material width for wavelengths 256 nm, 384 nm, and 770 nm. The bandwidth to wavelength ratio equals 0.39%. The initial pulse duration is 120 fs based on the estimated pulse duration from the tripler.

Chromatic dispersion made experimental measurement more difficult, because as the pulse duration increased, the irradiance was reduced, and the amount of two-photon absorbance decreased, as shown in Figure 9.
Figure 9. Theoretical two-photon absorbance versus xenon number density for various pulse durations.

In the experimental setup (Figure 3) the optics before the location of the point measurement were lens, L1, and the first test cell window. The center thickness of L1 (Thorlabs, LA4725) was 4.4 mm, and the test cell window had a thickness of 25.4 mm; therefore, the total UV fused silica material thickness was 29.8 mm for the optics before the point measurement. Figure 8 shows, for a total thickness of 29.8 mm, the pulse duration of the 256 nm laser beam increased to 308.4 fs. This pulse duration will be used throughout the paper for calculating the two-photon absorbance and beta measurements. Note that the test cell window was designed for a maximum allowable pressure of 17 MPa; therefore, in future experiments the windows could be designed with a smaller thickness to have less effect on pulse duration.
5.2 Spherical Aberrations

Another challenge working in the UV range is lens selection. Spherical lenses do not generally produce diffraction limited-spot sizes as a result of spherical aberrations. Aspheric lenses are designed to reduce spherical aberrations; however, they are mainly made of molded glass which has poor transmission in the UV (~0% at 256 nm). As a result, only spherical lenses are available since they use materials suited for high UV transmission. To achieve the highest amount of two-photon absorption a $M^2$ value of 1 is ideal (Section 4.2) which implies a diffraction-limited spot size. Therefore, it is important to reduce spherical aberrations and to optimize the focusing lens, L1, inside the test cell (Figure 3).

To reduce spherical aberrations, a longer focal length lens was preferred. This result was shown by using ZEMAX Optical Design Program to model lenses with various focal lengths. ZEMAX modeled the laser beam using a physical optics propagation which accounts for diffraction. For a 6mm input beam diameter (estimated tripler output diameter in the x-direction), the real focused spot size, $\omega_{\text{real}}$, was determined for a 50 mm, 75 mm, and 100 mm focal length lens. Note, the tripler output has an elliptical shape. The larger beam diameter of 6 mm (x-direction) was used for the ZEMAX ray trace analysis since the effect on the real beam waist caused by spherical aberrations is greater for larger beam diameters (see Appendix III b, Figure 26). To compare the real spot size to the diffraction limited spot size, the $M^2$ factor was calculated for each lens from Equation 12 a. in Section 4.2. Table 1 shows as the focal length increased the real
spot size, $\omega_{\text{real}}$, was closer to the diffraction limited spot size, $\omega_0$; therefore, a longer focal length was preferred to reduce spherical aberrations.

<table>
<thead>
<tr>
<th>UV Fused Silica Plano-convex Lenses</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thorlab #</td>
</tr>
<tr>
<td>LA4148</td>
</tr>
<tr>
<td>LA4725</td>
</tr>
<tr>
<td>LA4380</td>
</tr>
</tbody>
</table>

Table 1. $M^2$ values caused by spherical aberrations for UV fused silica plano-convex lenses at different focal lengths.

Although the optimum lens would be the longest focal length, the Thorlabs lens LA4725 with a 75 mm focal length was selected for the focusing lens, L1, in the experimental setup due to size constraints of the test cell. This was the same lens as used for the determination of the experimental $M^2$ value, as outlined in Section 4.3. For future experiments L1 could be placed outside the test cell; however, due to the test cell large window thickness of 25.4 mm the lens was placed inside the test cell for optimum focusing.
6. RESULTS

6.1 Theoretical and Measured Comparison

The test cell at room temperature was pressurized from 0 to 448 kPa with xenon gas. At pressure increments the $3\omega$ beam power was recorded before and after the test cell. At a pressure of 0 kPa the initial and final power measured to be 49.4 mW and 33.8 mW. As a result, the overall transmitted power through the lenses, L1 and L2, and the test cell windows was 68%. This corresponded to 8% less transmitted power for the optical path than expected for Fresnel losses [20] only. The transmission loss could be a result of unclean optics, and the uncertainty in the measurement of window fouling, which will be discussed in Section 7.2. At each pressure increment, the initial irradiance, $I_0$, and the irradiance over the absorption length, $I_\lambda$, (labeled in Figure 3) was calculated based on the transmission loss at a pressure of 0 kPa, $M^2$ value of 3.639, pulse duration of 308 fs, and beam waist of 4.8 $\mu$m. The absorption length, $l$, for this experiment based on the beam waist was 0.565 mm. Initial irradiance, $I_0$, varied in the experiment by 0.41%, which was most likely a result of the optical alignment drifting throughout the setup or degradation of the tripling crystal. Lastly, the measured two-photon absorbance and beta were calculated for each pressure increment. The results will first be presented in terms of a more familiar parameter, two-photon absorbance, and then the results will be presented in terms of the more suitable parameter, beta. Lastly, the uncertainty of temperature will be analyzed for the beta parameter.
The measured two-photon absorbance versus xenon number density was plotted along with the theoretical result as shown in Figure 10. The theoretical two-photon absorbance was plotted based on the measured $I_0$ at a pressure of 0 kPa, a xenon mole fraction of 1, and an absorption cross-section of $4.0 \times 10^{-43}$ m$^4$.

![Figure 10. Comparison between the theoretical and measured two-photon absorbance versus xenon number density. The theoretical is plotted for $M^2$ value equal to 3.639 and 2.59. The measured results are plotted for pure xenon and pure nitrogen in the test cell separately.](image)

At a total pressure of 448 kPa in the test cell, there was a measured 8.04% two-photon absorbance of xenon, where as, the theoretical code predicted 1.4 times less two-photon absorbance. Although there were discrepancies for the absolute values between the measured and predicted two-photon absorbance, the trend was similar. By reducing the $M^2$ value to 2.59, the measured results closely agreed to the theoretical curve as shown in Figure 10. Also, considering this is only a simplistic theoretical model, for instance, a top hat radial and temporal beam profile was assumed, the theoretical and measured
results fit well. Nevertheless, the major sources of bias were the code assumptions and the uncertainty of code parameters, such as $M^2$. The results for beta will consider the uncertainty of the absorption cross-section code parameter as well as the uncertainty of measurement from the resolution of the power meter.

To verify the measured two-photon absorbance was a result of xenon inside the test cell, the same experiment was preformed with nitrogen gas only. With nitrogen gas only, the average transmitted power for the test cell was 68.36%, similar to the results at a pressure of 0 kPa for the xenon results. Two-photon absorbance and beta for each increment was calculated based on the average transmission loss, and calculated similar to the xenon test. Ideally, as the test cell was pressurized from 0 to 448 kPa with nitrogen gas only, the amount of two-photon absorbance versus xenon number density should equal to zero. As projected, Figure 10 shows the average two-photon absorbance with nitrogen is $6.5 \times 10^{-7}$.

Beta is a more useful parameter since it is not dependent on initial irradiance and it is directly proportional to xenon number density. Therefore, beta was used for comparison between the measured and theoretical results. The measured and theoretical results of beta versus xenon number density were plotted in Figure 11. Beta is a linear function of xenon number density, and the measured beta showed this linear relationship as well.
Theoretical vs Measured Beta

Figure 11. Comparison between theoretical and measured beta versus xenon number density. The theoretical beta was plotted for the absorption cross-section, $\sigma = 4 \times 10^{-43} \text{ m}^4$, the upper uncertainty limit, $\sigma = 6 \times 10^{-43} \text{ m}^4$, and $\sigma = 5.6 \times 10^{-43} \text{ m}^4$. The measured results are plotted for pure xenon and pure nitrogen in the test cell separately, and the error bars represent uncertainty of beta.

The theoretical beta is dependent on the absorption cross-section. Reference [12] states an absorption cross-section, $\sigma$, equal to $4.0 \times 10^{-43} \text{ m}^4$; however, there is a stated uncertainty of 50%. Figure 11 shows the theoretical beta for the upper uncertainty limit of the absorption cross-section equal to $6.0 \times 10^{-43} \text{ m}^4$, and the measured beta fits within this uncertainty. For this experiment, the absorption cross-section was found to be $5.6 \times 10^{-43} \text{ m}^4$ for the measured results of beta. The error bars on the measured results for the xenon and nitrogen test represent uncertainty of beta discussed in the next section.
6.2 Uncertainty of Temperature

Beta is a function of temperature, and to measure the uncertainty of temperature, the uncertainty of beta has to be known. The uncertainty of beta was derived based on reference [21] as shown in Equation 13.

\[ u(\beta)^2 = \frac{1}{I_i} u(I_i)^2 + \frac{1}{I_0} u(I_0)^2 \] (13)

Where, \( u(I_i) \) = Uncertainty of the irradiance over the absorption length, \( I_i \)

\( u(I_0) \) = Uncertainty of the initial irradiance, \( I_0 \)

The Melles Griot power meter resolution equaled ±0.05 mW, as a result of the digits on the power meter display. From the power meter resolution, a \( M^2 \) value of 3.639, pulse duration of 308 fs, and beam waist of 4.8 \( \mu \)m, the uncertainty of \( I_i \) and \( I_0 \) equaled 7.694 x \( 10^9 \) W/m\(^2\). Figure 11 error bars on the measured results represent the associated uncertainty of beta.

From the ideal gas law (Equation 11 in Section 4.1) and beta parameter (Equation 10 in Section 4.1), temperature is derived to be a function of beta shown in Equation 14.

\[ T = \left( \frac{2l\sigma G_C}{h\gamma \nu_i} \right) \frac{X_{Xe}P}{k\beta} \] (14)

From Equation 14, the uncertainty of temperature is derived based on reference [21]. The result is shown in Equation 15. Therefore, the uncertainty of temperature is a function of the uncertainty of beta (Equation 13) as well as the parameter beta.

\[ u(T) = T \frac{u(\beta)}{\beta} \] (15)
The percent uncertainties of temperature for the measured results were plotted in Figure 12. At the highest pressure of 448 kPa, the uncertainty of temperature equaled 2.3%. To reach an uncertainty of temperature of 1%, based on the curve, an estimated pressure of 1.1 MPa is needed for the test environment.

Figure 12. Temperature uncertainty versus xenon number density for the measured results of beta.
7. LIMITATIONS ON APPLICATION

7.1 Introduction to Window Fouling

For calculating the measured initial irradiance, $I_0$, and the irradiance over the absorption length, $I_a$ (Figure 3) the assumption was made that the optics before and after the point measurement had equal transmission loss. However, two-photon absorption is a non-linear process and different results will occur depending on whether the transmitted intensity decreases before or after the measurement point.

As the optical setup in Figure 3 shows, the power was recorded before the first test cell window and the focusing lens, L1, at which point the beam was absorbed by the xenon gas at the point measurement location. After the beam was absorbed, it propagated through the second test cell window and the focusing lens, L2, before the final power was recorded. The overall transmission loss was determined for the entire optical path (68% transmitted power at 0 kPa given in Section 6.1). Accordingly, it was impossible to experimentally determine if the transmission loss occurred before or after the point measurement. Therefore, it was assumed there was an equal transmission loss before and after the point measurement to calculate $I_0$ and $I_a$ (labeled on Figure 3).

To simplify the explanation, consider only the test cell windows before and after the point measurement as shown in Figure 13. The first window the beam propagates through is $W_1$ and the second window is $W_2$. Similar to the experimental setup (Figure 3), the irradiance of beam will be measured before and after the optics, labeled $I_1$ and $I_2$.
in Figure 13, and their values will be used to calculate the total transmission loss, $T_{total}$, for the optics. Assuming equal transmission loss for the optics, the initial irradiance, $I_0$, and the irradiance over the absorption length, $I_l$, is calculated as shown in equations below.

$$I_0 = I_1 \sqrt{T_{total}} \quad (16 \ a)$$

$$I_l = I_2 \sqrt{T_{total}} \quad (16 \ b)$$

There is experimental error associated with the assumption of equal transmission loss (Equations 16a and 16b). The theoretical code was used to illustrate this error by examining the two extreme cases: 1) “Upstream loss” and 2) “Downstream loss.” For the upstream loss, shown in Figure 13(a), the first window, W1, is dirty causing the entire transmission loss ($I_0 = I_1 * T_{total}$) and the second window, W2 is assumed to be ideal with no transmission loss ($I_l = I_2$). The reverse case is the downstream loss, and this case models W1 as ideal with no transmission loss ($I_0 = I_1$) and W2 as dirty causing the entire transmission loss ($I_l = I_2 * T_{total}$).

Figure 13. Extreme fouling window cases: (a) Upstream loss and (b) Downstream loss.
To demonstrate the different results for these two extreme cases of window fouling, the initial irradiance, $I_0$, and the irradiance over the absorption length, $I_l$, was plotted versus total transmission for each case shown in Figure 14. For the upstream loss case, $I_0$ is a function of $T_{total}$. In contrast, the initial irradiance, $I_0$, for the downstream case is a constant value since it does not depend on total transmission. The plot was constructed for the following code parameters: initial average power of 100 mW, pulse duration of 308 fs, pure xenon, $M^2$ of 1, and a beam waist of 4.8 µm.

Two-photon absorption is a non-linear process dependent on the initial laser irradiance. Accordingly, as the initial irradiance decreases for the upstream case, there is less two-photon absorption of xenon and a higher irradiance, $I_l$. Whereas, the downstream case, is an after-result not changing the initial irradiance, $I_0$, and not changing the amount of two-photon absorption of xenon; however, the irradiance, $I_l$, will decrease due to the transmission loss at the window, $W_2$. Also, note the irradiance, $I_l$, for the upstream case is a non-linear function of $T_{total}$ while the irradiance, $I_l$, for the downstream case is a linear function of $T_{total}$.
7.2 Systematic Error due to Window Fouling

One of the main drawbacks unique to this optical technique was error caused by window fouling (introduced in Section 7.1). For the measured results described in Section 6.1, the total transmission for the optical setup was calculated once at a pressure of 0 kPa; therefore, it was assumed the total transmission did not change over the experiment. To apply this technique to systems of interest (Section 11.2 proposes measurement in a vortex tube), another method for measuring the total transmission, $T_{total}$, was needed. This method utilized a second beam. To measure changes in $T_{total}$, the second beam must travel along the same optical path and must not participate in the two-photon transition. Therefore, this beam could be at a non-resonant wavelength, a weaker average power, or a longer pulse duration. The proposed setup for the vortex tube used the discarded $2\omega$ beam from the tripler in the optical setup in Figure 3 since it was readily available and at
a non-resonant color for xenon. However, it is still necessary to assume that the windows are equally balanced.

To determine the systematic error based on this assumption a new parameter, $\varepsilon$, was defined. If $\varepsilon$ is equal to zero, the windows are equally balanced, which represents the experimental assumption. If $\varepsilon$ is equal to 1, the windows have complete unbalance, modeling the extreme cases plotted in Figure 14.

The following equations show the relationship for $I_0$ and $I_1$ based on total transmission measured by the $2\omega$ beam (proposed in Section 11.2.) and the unbalance parameter, $\varepsilon$.

$$I_1 = I_2 \left( T_{\text{total}} \right)^{\frac{1-\varepsilon}{2}} \quad (17 \ a)$$

$$I_0 = I_1 \left( T_{\text{total}} \right)^{\frac{1+\varepsilon}{2}} \quad (17 \ b)$$

$I_1$ and $I_2$ are considered the measured irradiance before and after the optics, for example, the irradiance locations labeled in Figure 13.

This optical technique monitored xenon number density; therefore, the parameter $\varepsilon$ was used to determine the error in measurement of xenon number density for the assumption of equally balanced windows. Figure 15 plots the ratio of the “real” xenon number density with unbalanced windows ($\varepsilon =$ equal increments of 0.111) to the “assumed” xenon number density with equally balanced windows ($\varepsilon = 0$) versus the total transmission measured by $2\omega$. The upper plot in Figure 15 shows the limits to stay within 10% error, of xenon number density measurement. For example, in the extreme
unbalance case of $\varepsilon = 1$, to maintain 10% error the total transmission must be greater than 81%. While with smaller $\varepsilon$, meaning windows of more similar transmission, the total transmission requirement for 10% error becomes lower.

A more common goal is to keep the error < 1%. Based on Figure 15, 1% error will generally require a high total transmission. At the extreme unbalance case, $\varepsilon = 1$, the total transmission must be greater than 97.8%, and at $\varepsilon = 0.111$, with close to equal window balance, the total transmission must be greater than 81.9%.

Figure 15. Systematic error due to window unbalance for various cases of $\varepsilon$ versus total transmission. $\varepsilon$ is incremented equally by 0.111 between 0 and 1. At $\varepsilon = 0$ the windows are equally balanced, and at $\varepsilon = 1$ the windows are completely unbalanced.
7.3 Beam Steering

As discussed in Sections 7.1 and 7.2, one drawback unique to this optical technique was window fouling. The second drawback for two-photon absorption is common to all optical techniques and it is known as beam steering. Nonlinear techniques such as presented here may be especially sensitive to beam steering. Beam steering is defined as the deviation of laser light from its intended path if gradients in index of refraction exist along the beam path. This definition and more detail on beam steering are found in reference [22]. For most systems, for example vortex tubes, pulse tubes, or internal combustion engines, there will be differences in index of refraction since it is dependent on changes in temperature, pressure, and composition of the gases present; therefore, as the beam propagates through these systems the intended path of a laser beam is altered. If the laser beam is focused inside one of these systems, as needed for two-photon absorption, the beam waist could unknowingly vary due to beam steering and change the \( M^2 \) value (Section 4.1) of the laser beam. This causes systematic error in measurement, because two-photon absorption is shown in Figure 6, Section 4.2 to decrease as the \( M^2 \) value increases.

Consider continuous point measurements of temperature are made across a vortex tube (proposed in Section 11.2) and gradients of index of refraction exist. It is experimentally difficult to determine which sections of the vortex tube will have more beam steering causing a larger \( M^2 \) value; therefore a constant \( M^2 \) value must be assumed for the entire vortex tube. This may result in large systematic error for measurement with this technique. Beam steering will cause fluctuations in \( M^2 \) value, generally resulting in
reduced two-photon absorption and will be incorrectly interpreted as reduced xenon number density.

Even with the assumption that the $M^2$ value is constant for the entire vortex tube, it is unknown how significantly beam steering will affect the factor $M^2$. However, initial studies from reference [23] conclude beam steering has a strong effect on $M^2$. These ongoing computations in our research group state an $M^2$ value as large as 36 for a turbulence model of an engine. However, two-photon absorption of xenon would still be possible with large $M^2$ values. The first generation optical setup (Appendix II) showed the theoretical code predicted an $M^2$ value of 29, and the measured two-photon absorbance was still detected at 1.5%. This was for an average laser power of 50 mW. Therefore, to measure two-photon absorption of xenon at higher $M^2$ values, higher laser beam irradiance would be required to achieve sufficient two-photon absorption. With the prospect of measuring higher $M^2$ values, a possible method to reduce the error caused by beam steering is to intentionally increase the $M^2$ value before it enters the device of interest. Therefore if the $M^2$ value is increased high enough, fluctuations in $M^2$ caused by beam steering in the device would become insignificant.
10. FLUORESCENCE RESULTS

10.1 Visual Indicator of Two-photon absorbance

As the 256 nm light was focused inside the test cell by the lens, L1, the beam fluoresced at the focus location, which could be seen by looking into the window perpendicular to the beam path. Atomic xenon fluorescence is in the infrared region; therefore, this fluorescence was only observed by the aid of an IR viewer. However, a bright green spot was observed just by looking into the test cell, and this fluorescence was assumed to be from a diatomic species. This “green” fluorescence was a nice feature of the two-photon absorption technique for alignment. There was a visual indicator of the point measurement location and verification that two-photon absorption was occurring inside the test environment. The fluorescence signal was recorded on a Thermal Oriel spectrograph and CCD camera with a long exposure time and accumulated over a 1000 runs. A signal was detected at center wavelengths of 878 nm, 900 nm, 978 nm, and 992 nm with a wavelength uncertainty of +/- 8 nm. The results are shown in Figure 16.
The reason four separate peaks were detected was unknown; however, a signal was expected at 905 nm for the two-photon excitation at 256 nm [12]. The fluorescence signal centered at 900 nm in Figure 16 was the strongest feature and most likely it was the fluorescence from the two-photon excitation at 256 nm. It was difficult to determine the remaining peak energy transitions, however, it was possible one of the fluorescence peaks was due to three photon ionization referred to as resonance enhanced multi photon ionization [(2+1) REMPI] [12]. Another spectrum which was unexpected was found around 540 nm, shown in Figure 17; however, this was probably the result of diatomic xenon or nitrogen fluorescence in the test cell. The more likely case was a nitrogen fluorescence, because there was a trace amount (<1%) of nitrogen in the test cell during this fluorescence measurement. However, further measurements are needed to verify this unknown spectrum.
Figure 17. Unknown spectrum from xenon fluorescence results. Most likely the result of diatomic nitrogen.
11. FUTURE WORK

Areas of future work include the study of reactive flows, continuous measurements along the flow path of the vortex tube and development of techniques for open systems. The technique of this project was developed primarily for two devices of non-reacting flows: vortex tube and pulse tube. The case study for measurement in a vortex tube was presented. This study did not include a pulse tube, but applying this technique to a pulse tube would be similar. Although two-photon absorption could be applied to reacting flows in a similar manner, this remains an area of future study. The proposed optical setup and method for making continuous point measurements of temperature are detailed in the following sections along with an investigation of different gases that could be used for open system measurements.

11.1 Measurement in a Vortex Tube

There is a need to further study continuous point measurements of temperature through the vortex tube. Figure 18 shows a schematic of the vortex tube which was built to allow optical access through the tube. The inlet port sends compressed gas tangentially to the tube creating a vortex. The gas is forced to one end of the tube where some of the gas escapes at a higher temperature (hot outlet port) while the remaining gas is forced back through the center of the tube. This inner stream gives off heat to the outer stream and exits at a colder temperature (cold outlet port). This energy separation between the inner and outer streams is not completely understood; therefore, continuous point measurements of temperature along the flow path of the vortex tube are desirable.
Two-photon absorption of xenon should provide successful results for measurement in the vortex tube; however, there are a few things to consider. First, this technique needs an optical path which allows a focus spot close to the diffraction limit (Section 4.2). Figure 18 is made of borosilicate glass which has low internal transmission in the UV (~0% at 256 nm). Therefore, a custom cylindrical vortex tube will be needed. Second, density gradients exist in the vortex tube causing beam steering that was described in Section 7.3 and further studies will be needed for determining the experimental error associated with beam steering for a vortex tube. Lastly, the vortex tube has a non-uniform pressure; therefore, direct measurement of the pressure will not be available. Since the pressure must be known to infer temperature with the two-photon absorption technique, the vortex tube will rely on pressure data from another method (either simulation code or another optical technique).

Figure 18. This vortex tube was built by Professor Greg Nellis at the University of Wisconsin-Madison.
11.2 Next Generation Optical Setup

The next generation optical setup is proposed to make continuous point measurement of temperature in the vortex tube using two-photon absorption of xenon. Figure 19 is a diagram of the proposed optical setup. This setup makes use of the doubled frequency, $2\omega$, from the tripler since it is a non-resonant color of xenon and readily available to deal with fluctuating laser intensity and to measure the total transmission simultaneously throughout the experiment. Therefore, this setup requires accurate and concurrent measurements at four different locations to measure two-photon absorption across the vortex tube.

![Figure 19. Next generation optical setup proposed for measurement in the vortex tube.](image)

Similar to the optical setup, Figure 3, the laser is a Spectra Physics Tsunami Ti:sapphire oscillator, and the output of the Ti:sapphire laser is sent through the TP-2000B fs tripler from U-Oplaz Technologies. However, this setup uses the combined light from the tripler, and the beam exiting the tripler contains the fundamental wavelength, $\omega$, the second harmonic, $2\omega$, and third harmonic, $3\omega$, generation. The fundamental wavelength,
ω, is filtered out by a custom CVI short pass filter. The remaining light, 2ω and 3ω, are used in the experiment, and this beam will be referred to as the main beam. Next, a sapphire beam splitter, Bs1, reflects 8% of the main beam in order to measure the initial intensity. The reflection from the beam splitter is sent through a high reflection mirror, M1, which reflects only 2ω and transmits the remaining 3ω light. The initial intensity of 2ω and 3ω are recorded on a high speed AXUV photodiode from International Radiation Detection (IRD), P3 and P1, respectively. (IRD photodiodes were demonstrated in the first generation optical setup, Appendix II). Continuing after the beam splitter, Bs1, the main beam is focused with an achromatic plano-convex lens, L1, into the vortex tube which provides the same focal length of 100 mm for 2ω and 3ω. L1 is placed on a translation stage to move the point measurement location determined by the focal length of L1 inside the vortex tube. The vortex tube is a cylindrical metal tube with a 25.4 mm outer diameter. An optical path will be provided by fitting two UV fused silica parallel windows on each side, each with a thickness of 6 mm and height of 12.7 mm. The working fluid in the vortex tube is xenon gas, which will absorb only the 3ω and not 2ω. After the vortex tube, the main beam is re-collimated by another achromatic plano-convex lens, L2, and propagates through the second beam splitter, Bs2, before the main beam is blocked. The reflection from Bs2 is sent through another high reflection mirror, M2, similar to the reflection path of Bs1. The final intensity of 2ω and 3ω are recorded on IRD photodiodes, P4 and P2, respectively.
11.3 Next Generation Method of Measurement

This setup has the advantage of taking continuous point measurements inside the vortex tube allowing simultaneous measurements at four different locations (P1, P2, P3, and P4). The measurement rate will be determined by the time response of the photodiodes. Since the $2\omega$ beam does not participate in the two-photon process, the transmission through the optical beam path is determined by photodiodes, P4 and P3. With the assumption the optics have an equal transmission before and after the point measurement, i.e. equally balanced windows, Equation 18 gives the transmission loss after L1 and the first vortex tube window.

\[ T_{\text{half}} = \frac{P_4}{\sqrt{P_3}} \quad (18) \]

Where $P_3$ = Initial average power for the $2\omega$ beam

$P_4$ = Final average power for the $2\omega$ beam

The initial irradiance, $I_0$, (labeled on Figure 19) before the point measurement location is determined by Equation 19.

\[ I_0 = \frac{P_1}{f A_c \Delta t} \sqrt{\frac{P_4}{P_3}} \quad (19) \]

Where, $P_1$ = Initial average power for the $3\omega$ beam

$f$ = Pulse repetition rate of laser

$A_c$ = Cross-sectional area of the beam at the point measurement

$\Delta t$ = Laser pulse duration

The irradiance over the absorption length, $I_\alpha$, (labeled on Figure 19) is determined by Equation 20.
\[ I_i = \frac{P2}{fA_\Delta t \sqrt{\frac{P4}{P3}}} \]  \hspace{1cm} (20)

Where \( P2 \) = Final average power for the \( 3\omega \) beam

Lastly, beta is calculated by Equation 21.

\[ \beta = \frac{1}{I_0} \left( \frac{I_0}{I_i} - 1 \right) = \frac{fA_\Delta t}{P1 \sqrt{\frac{P4}{P3}}} \left( \frac{P1}{P4} \frac{P4}{P2} \frac{P2}{P3} - 1 \right) \]  \hspace{1cm} (21)

While the next generation optical setup in Figure 19 shows measurements are required at four photodiodes to obtain the average initial and final power for the \( 2\omega \) and \( 3\omega \) beams (P1, P2, P3, and P4), Equation 21 shows beta is determined by only three signals: the ratio of the \( 3\omega \) beam power (P1/P2), the ratio of the \( 2\omega \) beam power (P4/P3), and the initial average power for \( 3\omega \), P1. To measure the power ratio signals, the next generation optical setup will use two balanced photoreceivers (New Focus 125-kHz Nirvana Auto-Balanced Photoreceiver). A balanced photoreceiver measures the ratio of two signals by canceling out the “common-mode” noise present in both signals. Therefore, each balanced photoreceiver will consist of two IRD photodiodes to collect the corresponding signals for either the \( 2\omega \) or \( 3\omega \) beam. The initial average power of \( 3\omega \), P1, will be determined along with the ratio from the same balanced photoreceiver.

Beta is directly proportional to xenon number density. After beta is measured for the point location in the vortex tube, xenon number density is determined from Figure 5, Section 4.1, and with known pressure, the temperature will be inferred from the ideal gas law, Equation 11 in Section 4.1.
11.4 Different Gases

Due to the high expense of xenon gas (xenon research grade, 25 volume liters at standard temperature and pressure, was approximately $200) the two-photon absorption technique is mostly limited to closed environments or systems with a small leak rate. To expand this technique for open systems, other tracer gases, such as argon and nitrogen, were examined.

Argon has a three-photon transition at 314.5 nm [24] which can be conveniently accessed with a frequency-doubled Ti:sapphire laser. For the laser line width (FWHM), $\Delta \nu_1 = 101$ cm$^{-1}$ (same line width for the experimental setup Figure 3), the three-photon excitation cross-section, $\sigma_{(3)}$, equals $1.141 \times 10^{-105}$ m$^6$s based on equations and constants provided elsewhere [25]. A theoretical code was written (Appendix IV MATLAB Files; f. Code 6) to calculate the three-photon absorbance based on equations derived by [23]. The following input code parameters were used: the laser line width and three-photon cross-section stated above, argon mole fraction of 1, pulse duration = 180 fs, cross-sectional area = 72 µm$^2$, convolution of the laser line profile = $3.49 \times 10^{-14}$ rad/s. The results found the three-photon absorbance of argon at 314.5 nm to be very weak. At an extremely high average power of 1 MW and a pressure of 689 kPa the three-photon absorbance equaled 0.0304%. The signal strength for other excited states of argon may be stronger. However, in order to determine the signal strength the multi-photon absorption cross-section is needed and in many cases it is not accurately known.
Molecular nitrogen has a two-photon transition at 283 nm [26]; however, the two-photon absorption cross-section is unknown and instead it is estimated based on a similar transition in OH [27]. With the assumed two-photon absorption cross-section and equations derived by [23], the two-photon absorbance of molecular nitrogen at 283 nm was calculated for the following input code parameters: nitrogen mole fraction of 1, pulse duration = 180 fs, cross-sectional area = 72 µm², line-resolved two-photon cross sections = $2 \times 10^{-41}$ m⁴/W (Appendix IV MATLAB Files; f. Code 6). The results for molecular nitrogen at 283 nm are stronger than argon presented above, but the signal strength is still weak. At an average power of 10 W and a pressure of 689 kPa the two-photon absorbance equaled 1.47%.

Compared to the three-photon transition of argon at 314.5 nm and the two-photon transition of molecular nitrogen at 283 nm, xenon has significantly higher signal strength for the two-photon transition at 256 nm. There is a possibility other multi-photon transitions may be stronger for argon or nitrogen; however, further analysis is needed to determine the absorption cross-sections, and, consequently, to expand this technique to a wider range of open and closed systems.
12. CONCLUSIONS

12.1 Summary of Results

This paper demonstrates a new spatially resolved optical technique to measure temperature using two-photon absorption of xenon by exciting the 256 nm two-photon transition of xenon inside a test cell at room temperature. A theoretical model was developed and compared to the measured results inside the test cell, and a close agreement was found despite the simplistic model of the code. The parameter beta was derived as the more suitable parameter for future two-photon absorption measurements since it is directly proportional to xenon number density and independent on initial irradiance. The uncertainty of temperature for this experiment was derived based on the uncertainty of the parameter beta. Also, the fluorescence at the focus of the laser beam inside the test cell was found to be a valuable tool for verifying the two-photon absorption transition. Lastly, a proposed optical setup and method for making continuous point measurements were theoretically developed for proposed future work.

12.2 Recommendations for Future Work

Two-photon absorption of xenon technique was developed specifically for measuring temperature inside a vortex tube or pulse tube. An optical setup and a method for making continuous point temperature measurements in a vortex tube are included in this paper. Although this technique was never directly applied to the vortex tube or pulse tube, two-photon absorption should prove successful for either of these devices. However, further
investigations are needed on the effect of window fouling and beam steering for future devices which are the main limitations of this technique.

Lastly, this technique should be expanded to a wider range of systems, including open systems and systems of reacting flows; therefore, two-photon absorption method with different gases or with a lower concentration of xenon should be examined. The initial studies for the multi-photon transitions of molecular nitrogen and argon found the signal strength to be extremely weak compared to the 256 nm transition of xenon; however, further research may lead to stronger multi-photon transitions. Another possibility is applying two-photon absorption with a lower xenon concentration; in general, a higher irradiance will be needed to achieve the same two-photon absorbance of xenon.
REFERENCES


APPENDIX

I. Experimental Optical Setup Pictures

The following pictures show the optical setup portrayed in Figure 3.

Figure 20. Experimental optical set up from right to left: Spectra Physics laser, filter, tripler, test cell.

Figure 20 shows the first steps of the optical setup. The Spectra Physics Tsunami laser produced 768 nm light (shown as red line), and the light emitted from the Tsunami was sent through a filter to attenuate the light. After the filter the laser beam entered the U-Oplaz Technologies tripler. The tripler generated the tripled frequency light, $3\omega$, at 256 nm (shown as blue line), and the light entered the test cell.
Figure 21 shows the test cell, the lens, L2, and the power meter. Inside the test cell, not visible, is the focusing lens, L1. The lens L1 was mounted in a lens holder which fit the 50.8 mm inner diameter of the test cell, and fixed within millimeters of the first test cell window. The focus location of L1 was visible by the window perpendicular to the beam path. As shown in Figure 21, the test cell was built by modifying a large Swagelok union tee (50.8 mm inner diameter). The test cell was constructed by drilling out the three end caps and sealing each end with an o-ring and a UV fused silica 25.4 mm thick window. A thickness of 25.4 mm was selected for design based on a maximum allowable pressure of 17 MPa. The test cell was placed on a two-axis translation stage for alignment with the beam path. The top flat surface of the test cell was fitted with an o-ring seal male thread (6.35 mm outer diameter tube connector) for connection of the xenon and nitrogen gas tanks as well as the pressure gauge.
II. First Generation Design

a. Optical Setup

Figure 22 is a schematic for one of the initial optical setups used to validate the two-photon absorption of xenon technique. This optical setup demonstrated the use of a non-resonant color, the doubled frequency, $2\omega$, of the tripler, to measure the changes in total transmission and laser intensity fluctuations.

![Diagram of the first generation optical setup. Demonstrates the use of a non-resonant color for measurement of the total transmitted intensity.](image)

This optical setup used the same laser and tripler from Figure 3; however, the main differences of this setup are the use of a prism and a shorter focal length of 20 mm inside the test cell. The laser is the same Spectra-Physics Tsunami Ti:sapphire oscillator with an 80 MHz repetition rate delivering an estimated 54 fs pulses at 768 nm with an average power of approximately 1.45 W and a FWHM of 15 nm. In order to generate a laser of wavelength 256 nm, the output of the Ti:sapphire laser is sent through a third-harmonic
generation (THG) package from U-Oplaz Technologies (TP-2000B fs tripler). The tripler produced $3\omega$ and $2\omega$ light at 256 nm and 384 nm, respectively, each with approximately 100 mW average power and 180 fs pulse duration. An iris was used to reduce the power of the $2\omega$ beam to equal the $3\omega$ beam power. The 256 nm and 384 nm light were made collinear by an equilateral dispersing prism (CVI # EDP-25.0-UV, 25 mm equilateral prism). A ZEMAX analysis for alignment of the prism is shown in Appendix III c. After the prism both beams were slowly focused by a bi-convex lens, L1, and traveled along the same path into the tee-cell. The test cell was a 50.8 mm inner diameter Swagelok union tee with UV fused silica windows and the cell was pressurized with xenon gas. Inside the test cell a plano-convex lens (Thorlabs LA4647), L2, with a 20 mm focal length was used to focus both beams. At the focus of L2 the $3\omega$ beam was absorbed by the xenon gas, whereas, the $2\omega$ beam was not absorbed; therefore, the $2\omega$ was used as a reference beam. Finally, both beams were refocused onto a high speed AXUV silicon photodiode from International Radiation Detection (IRD) by a plano-convex lens, L3. The IRD photodiode (part #AXUVHS5, 700 ps rise time, sensitive area of 1 mm$^2$) voltage output was recorded on an oscilloscope. To record the voltage, the beams were blocked individually by a chopper wheel. The chopper wheel was located at the exit of the tripler and set at a frequency of 420 Hz.

b. Results

The test cell was pressurized from 276 kPa to 931 kPa with xenon gas. At approximately 69 kPa increments the $3\omega$ and $2\omega$ beams voltage were recorded separately from an oscilloscope. Since the $2\omega$ beam does not participate in the two-photon absorption, this
beam was recorded to measure the transmission loss and laser intensity fluctuations. The $3\omega$ beam voltage was adjusted accordingly for changes in $2\omega$, and the two-photon absorbance at each increment was determined and plotted versus xenon number density for the $3\omega$, shown in Figure 23.

![Figure 23. Measured two-photon absorbance of xenon for the first generation optical setup. The error bars represent uncertainty of two-photon absorbance.](image)

There was a measurement uncertainty of 0.4 mV for the oscilloscope which corresponds to an absorbance uncertainty of 0.0794%, represented by error bars on Figure 23. For estimating the uncertainty of temperature it was assumed the measured results fit a linear line. This is a major assumption since Figure 4 in Section 4.1 shows two-photon absorbance is a non-linear function of xenon number density; however, it is useful to assume a linear fit to determine a rough estimate on the uncertainty of temperature. From
the slope of the fitted line, the uncertainty of absorbance can be related to \(~10.96\%\) uncertainty of temperature.

The measured two-photon absorbance was compared to the theoretical results as shown in Figure 24. The theoretical two-photon absorbance was plotted based on an average power of 50 mW, a $M^2$ value of 3.639, a xenon mole fraction of 1, an absorption cross-section of $4.0 \times 10^{-43} \text{ m}^4$, and pulse duration of 400 fs. Based on these input parameters, the code predicts 7 times more absorbance than experimentally measured.

This was one of the first optical setups to validate the two-photon absorption of xenon method, and although there are large differences between the absolute values of the measured and predicted two-photon absorbance, the trend was similar. By increasing the $M^2$ value to 29.09, the measured results show a similar curve, see Figure 24.
The main reason for the large predicted $M^2$ value is this optical setup uses a shorter focal length lens of 20 mm for the focusing lens L2 inside the test cell (Figure 22). As shown in Table 1 Section 5.2, the $M^2$ value for a focal length of 20 mm will be greater than 21 which was the $M^2$ value calculated for focal length equaled to 50 mm.

Also, the polarization of the $3\omega$ beam exiting the tripler was vertically polarized (based on TP-2000B fs tripler manufacture data), which caused large reflection loss from the prism. The prism was designed for horizontally polarized beams. Therefore, the average power may be lower than the modeled 50 mW. Along with the wrong polarization as the main beam was propagated through the prism, the beam became a little distorted. This could indicate an increase in $M^2$ through the prism. Lastly, with the addition of the prism and bi-convex lens, L1 (Figure 22) a longer pulse duration exists for this experiment, and
as shown in Figure 9 Section 5.1, a longer pulse duration decreases the two-photon absorbance.
III. ZEMAX Optical Design Program

a. Diffraction-limited Beam Waist

The diffraction-limited beam waist, $\omega_0$, is calculated based on Gaussian parameters shown in Equation 22 [28].

$$\omega_0 = \frac{\lambda}{\pi \tan(\theta_{\text{half}})}$$  \hspace{1cm} (22)

The half angle of divergence, $\theta_{\text{half}}$, is defined at the $\frac{1}{e^2}$ for intensity. To estimate the half angle of divergence for the experimental setup (Figure 3), ZEMAX Optical Design Program was used to perform a ray trace as shown in Figure 25. With the laser beam diameter of 2.3 mm and the plano-convex focusing lens (same as optical setup, Thorlabs LA4725), the estimated half angle of divergence is 0.96 deg. Using Equation 22 at a wavelength, $\lambda$, of 256 nm, the diffraction-limited beam waist, $\omega_0$, equaled 4.8µm.

![Figure 25. Ray trace using ZEMAX Optical Design Program to estimate the diffraction-limited beam waist, $\omega_0$, for the experimental setup.](image-url)
The laser for the experiment setup (Figure 3) is the frequency tripled, $3\omega$, light from the tripler unit. While the paper assumes a circular beam at a diameter of 2.3 mm, the $3\omega$ beam has an elliptical spot profile (y-direction diameter $\sim$ 2.3 mm and x-direction diameter $\sim$ 6 mm). A similar ray trace was performed for the x-direction with a beam diameter of 6 mm. With a larger beam diameter, the half angle of divergence is larger and ZEMAX calculates the half angle equal to 2.5 deg. Thus, the diffraction-limited beam waist, $\omega_0$, is smaller and is equal to 1.9 µm. The paper assumes a diffraction-limited beam waist equal to 4.8 µm from the y-direction since it will have the largest negative effect on the measured results for two-photon absorbance (see Section 4.1).

**b. Spot Size Caused by Spherical Aberrations**

ZEMAX Optical Design Program was used to determine the effect on the real beam waist, $\omega_{\text{real}}$, caused by spherical aberrations. A ray trace was performed for Thorlabs lens LA4725 (same lens as optical setup Figure 3) at various input beam diameters. ZEMAX calculates the real beam waist based on a physical optics propagation which accounts for diffraction, and the diffraction-limited beam waist, $\omega_0$, was calculated based on the ray trace half angle of divergence (Equation 22, Appendix III a.). Figure 26 shows the real beam waist and diffraction-limited beam waist was plotted for input beam diameters between 2 mm and 6.5 mm at increments of 0.5 mm.
Figure 26. The diffraction-limited beam waist, $\omega_0$, and real beam waist, $\omega_{\text{real}}$, for Thorlabs lens LA4725 at various input beam diameters.

There is a minimum real beam waist at an input beam diameter of 5 mm. This is considered the optimum performance point where spherical aberrations and diffraction form a minimum. However, at smaller input beam diameters the real beam waist closely agrees to the diffraction-limited beam waist (at 2 mm input beam diameter, $M^2 \sim 1$) even though the real beam waist is considerably larger. Therefore, spherical aberrations have a greater effect on $M^2$ value at larger input beam diameters.
c. Equilateral Dispersing Prism

The CVI prism used in the first generation optical setup (Appendix II a., Figure 22) is a 25 mm UV fused silica equilateral dispersing prism. To determine the proper alignment of the prism, ZEMAX Optical Design Program was used to model the CVI prism.

Figure 27 shows the prism’s entering and exiting angles, $\theta_1$ and $\theta_2$, respectively. The entering angle, $\theta_1$, is referenced normal to the prism face, and the exiting angle, $\theta_2$, is referenced from the entering line.

![Equilateral Dispersing Prism](image)

*Figure 27. Equilateral Dispersing Prism, $\theta_1$ is the entering angle (referenced to prism normal) and $\theta_2$ is the exiting angle (referenced to the line of the entering beam)*

By changing the entering angles of the modeled prism, ZEMAX was used to calculate the corresponding exit angles with respect to wavelength. The results are shown in Figure 28.
Exiting Prism Angle (Theta2) vs Wavelength for Various Entering Prism Angles (Theta1)

Figure 28. Exiting Prism angle, $\theta_2$, versus wavelength for various entering prism angles, $\theta_1$. 
IV. Bandwidth of the Laser Line

The spectral full width at half maximum (FWHM) was determined for the THG output of the tripler. The input beam to the tripler was fixed at a wavelength of 768 nm. By collecting the frequency tripled, $3\omega$, light into a 400 µm fiber, the spectrum was recorded on the Thermo Oriel spectrograph. Figure 29 shows the THG output has a FWHM of 1.03 nm with a peak wavelength of 252 nm. A Oriel calibration lamp, 6035 Hg(Ar), was used to calibrate the spectrograph at the wavelength of 253.65 nm and the spectrograph error was found to be +3.95. Therefore, the THG output is estimated to be centered at 255.95 nm (~256 nm). For a measured FWHM of 1 nm and a wavelength of 256 nm, the spectral full width at half maximum for the laser line is equal to 15258.8 m$^{-1}$.

Figure 29. The spectral full width at half maximum (FWHM) for the frequency tripled, $3\omega$, output of the tripler.
V. Tsunami Gaussian Fit

The radial distribution of the Spectra Physics Tsunami Ti:sapphire laser used in the experimental setup (Figure 3) was measured and compared to a Gaussian fit.

Figure 30. X-axis radial distribution of the Spectra Physics Tsunami Ti: sapphire laser.

Figure 30 shows the radial distribution in the x-direction which was along the axis parallel to the optical table. The measured 1/e² diameter in the x-direction was 1.26 mm. Similarly, the radial distribution for the y-axis was measured in Figure 31 and the 1/e² diameter was found to be 0.78 mm. The y-axis was defined as the axis perpendicular to the optical table. For the x and y axes, the radial distribution closely agrees to the Gaussian curve.

Also, note the beam was found to be elliptical with the x-direction as the largest 1/e² diameter. As the Ti:sapphire laser was sent through the tripler unit, the ellipticity was probably amplified which explained the large difference between the x and y axis for the 3ω beam profile.
Figure 31. Y-axis radial distribution of the Spectra Physics Tsunami Ti:sapphire laser.
VI. MATLAB Files

a. Code 1 (Theory.m)

```matlab
clc
clear all
close all

i=1;
j=50; %increment in pressure for plots

x=1.0; %xenon mole fraction; pure xenon assumed
P_max=100/14.7; % maximum total pressure [atm]
P_low=1e-1000/14.7; %minimum total pressure [atm]
P=[P_low:(P_max-P_low)/(j-1):P_max]; %matrix for total pressure [atm]
k=1.38e-23; % Boltzmann constant [J/K]%
T=298; %Temperature [K]%
n=x.*P.*1.01325e5./(k.*T); %number density of Xe [atoms/m^3]%

wavelength=256e-9; %wavelength [m]%
sigma=4e-43; %xenon absorption cross-section [m^4]%
h=6.626e-34; %Planck's constant [J-s]%
c=3e8; %speed of light in a vacuum [m/s]%
v=1/wavelength; %laser wavenumber [m^-1]%
G=1; %second-order coherence function of the photon field%
Freq=80e6; %Repetition rate of laser [Hz]%
Delta_v_laser=15258.8; %FWHM of laser line, assumes Fourier-limited pulse [m^-1]%
Delta_v_Xe=0*(100); %[m^-1] absorption line FWHM (Neglected) %
C=sqrt((4*log(2))/(pi*(2*Delta_v_laser^2+Delta_v_Xe^2))); %convolution assuming gaussian [m]%
Pulse_dur=308.4e-15; %pulse duration [sec]%

M2_max=5; %maximum M^2 value
M2_low=1; %minimum M^2 value
M2=[M2_low:(M2_max-M2_low)/(j-1):M2_max]'; %matrix for M^2 values
w_0=4.8; %diffraction limited beam waist, calc from ZEMAX [um]%
w_real=w_0.*((M2).^0.5).*10^-6; %real beam waist [m]%
L=.000566 %absorption length [m] (does not depend on M^2 value)
A_c=pi.*w_real.^2; %Cross-sectional area of beam [m^2]%

Power=100*10^-3; %Initial average power [W]%
T_max=1; %max transmission [%]%
T_min=0; %min transmission [%]%
T=[T_min:(T_max-T_min)/(j-1):T_max]'; %Transmission matrix%
```
\[ \text{Power}_T = \text{Power} \times T; \]  
% initial average power with varying transmission loss [W]%

I_0 = (\text{Power} / \text{Freq}) / (\text{Pulse}_dur \times A_c);  
% initial intensity [W/m}^2] (matrix varies M^2)%

I_0_T = (\text{Power}_T / \text{Freq}) / (\text{Pulse}_dur \times A_c(1,1));  
% initial intensity [W/m}^2] when varying Transmission%

% creates matrix for multiplication%
while i<=j
    I_0_2(1:j,i)=I_0(1:j,1);  
    I_0_T_2(1:j,i)=I_0_T(1:j,1);  
    i=i+1;
end

L_0 = (h \times c^2 \times v) / ((2 \times \sigma \times G \times C) \times (I_0 \times n));  
% length at 50% initial irradiance [m]%

L_0_T = (h \times c^2 \times v) / ((2 \times \sigma \times G \times C) \times (I_0_T \times n));  
% length at 50% initial irradiance [m] when varying transmission%

I_L = I_0_2 ./ (1 + L ./ L_0);  
% intensity over absorption length [W/m}^2]%

% Figure plots the theoretical absorbance versus xenon number density for
% for various M^2 values
figure
plot(n, absorbance,'-k'), xlabel('Xenon Number Density [atoms/m}^3]'), ylabel('Two-photon Absorbance'), grid on, legend('M^2=1','M^2=2','M^2=3','M^2=4','M^2=5'), title('Two-photon Absorbance of Xenon'), axis([n(1,1) n(1,j) 0 absorbance(1,j)])
% the M^2 value equal to 1
figure
plot(n,absorbance(1,:),'-k'),...
xlabel('\fontsize{12}Xenon Number Density [atoms/m^3]'),ylabel('\fontsize{12}Two-photon Absorbance'),...
grid on, legend('\fontsize{12} P=100mW, Pure xenon, \DeltaT=308fs'),...
title('\fontsize{12}Theoretical Two-photon Absorbance of Xenon'),
axis([n(1,1) n(1,j) 0 absorbance(1,j)])

%Figure plots the extreme cases for window fouling: "upstream loss" and
"downstream loss" for the irradiance over the absorption length
figure
plot(T,I_0_T,'k',T(j,1),I_0_T(j,1),'*',T,I_L_T(:,j),'k:',T,I_L_back,'b'),xlabel('\fontsize{12}Total Transmission [%]'),...
ylabel('\fontsize{12}Irradiance [W/m^2]'),...
title('\fontsize{12}Window Fouling'),...
legend('\fontsize{12}I_0 Upstream loss','\fontsize{12}I_0 Downstream loss','\fontsize{12}I_L Upstream Loss','\fontsize{12}I_L Downstream loss'),...
axis([T_min T_max I_0_T(1,1) I_0_T_2(j,j)])

b. Code 2 (Results.m)

% Matlab Code 2

%Results Section 6.1:  Figure 10, Figure 11, and Figure 12

clc
clear all
close all

i=1;
j=50; %increment for plots
%j=7 % used for calc error

x=1.0; %xenon mole fraction; pure xenon assumed
P_max=70/14.7; %maximum total pressure [atm]
P_low=1e-1000/14.7; %minimum total pressure [atm]
P=[P_low:(P_max-P_low)/(j-1):P_max]; %matrix for total pressure [atm]
%P=[12.5 20 30 40 50 60 65]./14.7; %used for calc mean error betw
theoretical and measured results
k=1.38e-23; %Boltzmann constant [J/K]%
T=298; %temperature [K]%
n=x.*P.*1.01325e5./(k.*T); %number density of Xe [atoms/m^3]%

wavelength=256e-9; %wavelength [m]%
sigma=4e-43; %xenon absorption cross-section [m^4]%
sigma_low=5.642e-43; %measured xenon absorption cross-section [m^4]%
sigma_high=6e-43; %upper uncertainty limit for xenon absorption cross-
section [m^4]%
h=6.626e-34; %Planck's constant [J-s]%
c=3e8; %speed of light in a vacuum [m/s]
v=1/wavelength; %laser wavenumber [m^-1]
G=1; %second-order coherence function of the photon field
FREQ=80e6; %Repetition rate of laser [Hz]
DELTA_V_LASER=15258.8; %FWHM of laser line, assumes Fourier-limited pulse [m^-1]
DELTA_V_XE=0*(100); %xenon absorption line FWHM (Neglected) [m^-1]
C=sqrt((4*log(2))/(pi*(2*DELTA_V_LASER^2+DELTA_V_XE^2))); %convolution assuming gaussian [m]

PULSE_MAX=308.4e-15; %maximum pulse duration [sec]
PULSE_MIN=100e-15;%minimum pulse duration [sec]
PULSE_DUR=[PULSE_MIN:(PULSE_MAX-PULSE_MIN)/(j-1):PULSE_MAX]'; %matrix for pulse duration [sec]

POWER=40.86*10^-3; %Initial average power [W]
M2=3.639; %experimental M^2 value
W_0=4.8; %diffraction limited beam waist, calc from ZEMAX [um]
W_REAL=W_0*sqrt(M2)*10^-6; %real beam waist [m]
L=2*pi*W_REAL^2/(wavelength.*M2); %absorption length [m]
A_C=pi*W_REAL^2 %Cross-sectional area of beam [m^2]

M2_2=2.59; %measured M2 for 2-photon absorbance plot
W_REAL_2=W_0*sqrt(M2_2)*10^-6; %real beam waist [m]
A_C_2=pi*W_REAL_2^2 %Cross-sectional area of beam [m^2]

I_0=(POWER/FREQ)./(PULSE_DUR.*A_C); %initial irradiance [W/m^2] (matrix varies Pulse_duration)
I_0_M=(POWER/FREQ)./(PULSE_DUR.*A_C_2); %initial irradiance [W/m^2] (matrix varies Pulse_duration)

while i<=j
  I_0_2(1:j,i)=I_0(1:j,1);
  I_0_M_2(1:j,i)=I_0_M(1:j,1);
  i=i+1;
end

L_0=(h*c^2*v)./((2*sigma*G*C).*(I_0*n)); %length at 50% initial irradiance [m] for M2=3.639%
L_0_2=(h*c^2*v)./((2*sigma*G*C).*(I_0_M*n)); %length at 50% initial irradiance [m] for M2=2.59%

I_L=I_0_2./(1+L./L_0); %intensity over absorption length [W/m^2] for M2=3.639%
I_L_2=I_0_M_2./(1+L./L_0_2); %intensity over absorption length [W/m^2] for M2=2.59%

abсорbance=log(I_0_2./I_L); %2-photon absorbance of xenon for M2=3.639%
abсорbance_2=log(I_0_M_2./I_L_2); %2-photon absorbance of xenon for M2=2.59%

BETA_2=(2*sigma*G*C.*n.*L)/(h*c^2*v); %theoretical beta [m^2/W] for sigma=4e-43%
\[
\text{Beta}_2\text{ high} = \frac{(2*\sigma_{\text{high}}*G*C*n*L)}{(\hbar*c^2*v)}; \quad \text{%theoretical beta [m^2/W for } \sigma_{\text{high}}=6e-43\% \\
\text{Beta}_2\text{ low} = \frac{(2*\sigma_{\text{low}}*G*C*n*L)}{(\hbar*c^2*v)}; \quad \text{%theoretical beta [m^2/W for } \sigma_{\text{low}}=5.642e-43\% \\
\]

% Measured results from Xenon Test
\[
P_{0\text{ avg}} = [40.77949046 \ 40.77949046 \ 40.69677344 \ 40.69677344 \ 40.69677344 \\
40.61405642 \ 40.61405642].*10^{-3}; \quad \%\text{average initial power [W]} \\
P_{L\text{ avg}} = [40.13684285 \ 39.77416053 \ 39.29058411 \ 38.80700769 \ 38.20253717 \\
37.83954853 \ 37.47717254].*10^{-3}; \quad \%\text{average final power [W]} \\
I_{0\text{ meas}} = P_{0\text{ avg}}./A_c./Freq./Pulse\_max; \quad \%\text{initial irradiance [W/m^2]} \\
I_{L\text{ meas}} = P_{L\text{ avg}}./A_c./Freq./Pulse\_max; \quad \%\text{irradiance over the absorption length [W/m^2]} \\
\text{absorbance meas} = \log(I_{0\text{ meas}}./I_{L\text{ meas}}); \quad \%2\text{-photon absorbance of xenon} \\
\text{Beta meas} = 1./I_{0\text{ meas}}.*(I_{0\text{ meas}}./I_{L\text{ meas}}-1); \quad \%\text{Measured beta} \\
p_{\text{meas}} = ([12.5 \ 20 \ 30 \ 40 \ 50 \ 60 \ 65]./14.7); \quad \%\text{total pressure increments [atm]} \\
n_{\text{meas}} = (p_{\text{meas}}.*1.01325e5./(k.*T)); \quad \%\text{xenon number density [atoms/m^3]} \\
\]

% Measured results from Nitrogen Test
\[
P_{0\text{ N2}} = [45.30915944 \ 44.97843565 \ 44.73039281 \ 44.56503091 \ 44.31698807 \\
44.15162617 \ 44.06894522 \ 44.06894522].*10^{-3}; \quad \%\text{average initial power [W]} \\
P_{L\text{ N2}} = [45.23412099 \ 44.99222729 \ 44.7503336 \ 44.62938675 \ 44.26654621 \\
44.14559936 \ 44.14559936 \ 44.02465251].*10^{-3}; \quad \%\text{average final power [W]} \\
I_{0\text{ N2}} = P_{0\text{ N2}}./A_c./Freq./Pulse\_max; \quad \%\text{initial irradiance [W/m^2]} \\
I_{L\text{ N2}} = P_{L\text{ N2}}./A_c./Freq./Pulse\_max; \quad \%\text{irradiance over the absorption length [W/m^2]} \\
\text{absorbance N2} = \log(I_{0\text{ N2}}./I_{L\text{ N2}}); \quad \%2\text{-photon absorbance of nitrogen} \\
\text{Beta N2} = 1./I_{0\text{ N2}}.*(I_{0\text{ N2}}./I_{L\text{ N2}}-1); \quad \%\text{Measured beta} \\
p_{\text{N2}} = ([0 \ 10 \ 20 \ 30 \ 40 \ 50 \ 60 \ 65]./14.7); \quad \%\text{total pressure increments [atm]} \\
n_{\text{N2}} = (p_{\text{N2}}.*1.01325e5./(k.*T)); \quad \%\text{xenon number density [atoms/m^3]} \\
\]

% Uncertainty Calculations for measured results
\[
P_{\text{meter uncert}} = 0.05e-3; \quad \%\text{uncertainty of the power meter [W]} \\
I_{\text{uncert}} = P_{\text{meter uncert}}./Freq./Pulse\_max./A_c; \quad \%\text{uncertainty of irradiance [W/m^2]} \\
\text{Beta uncert} = (1./I_{L\text{ meas}}.^4.*I_{\text{uncert}}^2+1./I_{0\text{ meas}}.^4.*I_{\text{uncert}}^2).^0.5; \quad \%\text{Beta uncertainty for xenon test} \\
\text{Beta uncert N2} = (1./I_{L\text{ N2}}.^4.*I_{\text{uncert}}^2+1./I_{0\text{ N2}}.^4.*I_{\text{uncert}}^2).^0.5; \quad \%\text{Beta uncertainty for nitrogen test} \\
T_{\text{per uncert}} = (\text{Beta uncert}./\text{Beta meas}).*100; \quad \%\text{percent temperature uncertainty for xenon test} \\
\]

\[
\text{Error} = \text{Beta}_2\text{ low}/\text{Beta meas}; \\
\%\text{Mean}=\text{mean}(\text{Error}); \\
\%\text{uncertainty}=\text{Error}/\text{Mean}; \\
\]

% Figure comparing the theoretical and measured beta versus xenon number density. 
% The theoretical beta plots the absorption cross section at sigma=[4e-43m^4 \ 6e-43m^4 \ 5.6e-43m^4]. 
% The measured results are plotted for xenon test and nitrogen test figure
plot(n,Beta_2_high',':k',n,Beta_2_low',':k',n,Beta_2,'k',n_meas,Beta_meas,'bo',n_N2,Beta_N2,'r*'),...
xlabel('$\text{Xenon Number Density}$
[atoms/m^3]'),ylabel('$\text{Beta}$
[m^2/W]'),...
title('$\text{Theoretical vs Measured Beta}$'),...
legend('$\sigma = 6.0e-43$ m^4','$\sigma = 5.6e-43$ m^4','$\sigma = 4.0e-43$ m^4','$\text{Measured Xe}$','$\text{Measured N2}$'),...
axis([n(1,1) n(1,j) -5e-16 Beta_2_high(1,j)])
errorbar(n_meas,Beta_meas,Beta_uncert,Beta_uncert,'ob')
errorbar(n_N2,Beta_N2,Beta_uncert_N2,Beta_uncert_N2,'r*')

%Figure plotting the temperature uncertainty versus xenon number
density
figure
plot(n_meas,T_per_uncert,'k-o'),xlabel('$\text{Xenon Number}$
Density [atoms/m^3]'),
ylabel('$\text{Temperature Uncertainty}$
[%]'),axis([n_meas(1,1) n_meas(1,7) 2 11.01]), grid on,
title('$\text{Temperature Measurement Uncertainty}$')

%Figure comparing the theoretical and measured two-photon absorbance
versus
%xenon number density. The theoretical results are plotted for
M^2=3.639
%and M^2=2.59. The measured results are plotted for xenon test and
%nitrogen test.
figure
plot(n,absorbance(j,:)','k',n,absorbance_2(j,:)',':k',n_meas,absorbance _meas,'bs',n_N2,absorbance_N2,'r*'),...
xlabel('$\text{Xenon Number Density}$
[atoms/m^3]'),ylabel('$\text{Two-photon Absorbance}$'),
legend('$\text{Theoretical}$
M^2=3.639','$\text{Theoretical}$
M^2=2.59','$\text{Measured Xe}$','$\text{Measured N2}$'),
title('$\text{Theoretical vs. Measured Absorbance}$'),axis([n(1,1) n(1,j) -.004 absorbance_2(j,j)])

c. Code 3 (Pulsedur.m)

%Matlab Code 3
%Chromatic Dispersion Plots
%Section 5.1: Figure 8 and Figure 9

clic
clc
clear all
close all

i=1;
j=5; %increment for plots

x=1.0; %xenon mole fraction, assume pure xenon%
\( P_{\text{max}} = 100/14.7; \) % max total pressure [atm]
\( P_{\text{low}} = 1e-100/14.7; \) % min total pressure [atm]
\( P = [P_{\text{low}}: (P_{\text{max}} - P_{\text{low}})/(j-1): P_{\text{max}}]; \) % matrix for total pressure [atm]
\( k = 1.38e-23; \) % Boltzmann constant [J/K]
\( T = 298; \) % temperature [K]
\( n = x.*P.*1.01325e5./(k.*T); \) % number density of Xe [atoms/m^3]

\( \text{wavelength} = 256e-9; \) % wavelength [m]
\( \text{sigma} = 4.0e-43; \) % [m^4] Xe absorption cross-section
\( \hbar = 6.626e-34; \) % Planck's constant [J-s]
\( c = 3e8; \) % [m/s] speed of light in a vacuum
\( v = 1/\text{wavelength}; \) % laser wavenumber [m^-1]
\( G = 1; \) % second-order coherence function of the photon field
\( \text{Freq} = 80e6; \) % Repetition rate of laser [Hz]
\( \Delta v_{\text{laser}} = 15258.8; \) % FWHM of laser line, assumes Fourier-limited pulse [m^-1]
\( \Delta v_{\text{Xe}} = 0*(100); \) % [m^-1] absorption line FWHM (Neglected)
\( C = \sqrt{(4*\log(2))/(pi^2*\Delta v_{\text{laser}}^2+\Delta v_{\text{Xe}}^2))}; \) % convolution assuming gaussian [m]

\( \text{Pulse\_max} = 500e-15; \) % max pulse durations [sec]
\( \text{Pulse\_min} = 100e-15; \) % min pulse dur [sec]
\( \text{Pulse\_dur} = [\text{Pulse\_min}: (\text{Pulse\_max}-\text{Pulse\_min})/(j-1): \text{Pulse\_max}]; \) % [sec]

\( M2 = 3.639; \) % experimental M^2 value
\( w_0 = 4.8; \) % diffraction limited beam waist, calc from ZEMAX [um]
\( w_{\text{real}} = \sqrt{\text{w}_0^2 + (2*M2)^2}; \) % real beam waist [m]
\( L = 2*pi*w_{\text{real}}^2/\text{wavelength}.*M2; \) % absorption length [m]
\( A_c = \text{pi}*w_{\text{real}}^2 \) % Cross-sectional area of beam [m^2]

\( \text{Power} = 100*10^-3; \) % initial average power [W]
\( I_0 = (\text{Power}/\text{Freq})./(\text{Pulse\_dur}*A_c); \) % initial intensity [W/m^2]

\% creates matrix for multiplication
\%while i<=j
\% I_0_2(1:j,i) = I_0(1:j,1); 
\% i = i + 1;
\%end

\( L_0 = (h*c^2*v)/(2*(\text{sigma}*G*C).*I_0*n); \) % length at 50% initial irradiance [m]
\( I_L = I_0_2./((1+L/L_0); \) % intensity over absorption length [W/m^2]
\( \text{absorbance} = \log(I_0_2./I_L); \) % two-photon absorbance of xenon

\%Plots the theoretical two-photon absorbance versus xenon number density
\%for various pulse durations
\%figure
\%plot(n,absorbance'), xlim('\text{Xe Number Density [Atoms/m^3]}'), ylim('\text{Two-photon Absorbance}'),
\% grid on, axis([\text{n}(1,1) \text{n}(1,j) 0 absorbance(1,j)]),
\% legend('\text{100fs}','\text{200fs}','\text{300fs}','\text{400fs}','\text{500fs}'))
title('Two-photon Absorbance for Various Pulse Durations');

BW=1.0; % FWHM of 256nm laser line [nm]%
BW_g=1.5; % FWHM of 384nm laser line [nm]%
BW_r=3; % FWHM of 770nm laser line [nm]
glass_width_pres=[0:1:100]; % [mm] matrix for total UV fused silica material
Pulse_dur_tripler=123.87e-15; % [sec] assumed initial pulse duration of beam
Dis_coeff_256=6151; % [ps/nm/km] dispersion coefficient of fused silica at 256nm%
Dis_coeff_770=122.53; % [ps/nm/km] dispersion coefficient of fused silica at 770nm%
Dis_coeff_384=1320.5; % [ps/nm/km] dispersion coefficient of fused silica at 384nm%
Pulse_dur_256=Pulse_dur_tripler+(Dis_coeff_256.*BW.*(glass_width_pres.*10.^-6).*10.^-12); % Pulse duration at 256nm [sec]%
Pulse_dur_770=Pulse_dur_tripler+(Dis_coeff_770.*BW_g.*(glass_width_pres.*10.^-6).*10.^-12); % Pulse duration at 770nm [sec]%
Pulse_dur_384=Pulse_dur_tripler+(Dis_coeff_384.*BW_r.*(glass_width_pres.*10.^-6).*10.^-12); % Pulse duration at 384nm [sec]%

% Plots the pulse duration versus UV fused silica material width for an initial pulse duration of 120 fs
figure
plot(glass_width_pres,Pulse_dur_256.*10^15,glass_width_pres,Pulse_dur_384.*10^15,glass_width_pres,Pulse_dur_770.*10^15),...
xlabel('
UV Fused Silica Material Width [mm]'),ylabel('
Pulse Duration [fs]'),...
title('Chromatic Dispersion for Initial Pulse Duration of 120fs'),...
legend('
256nm','384nm','770nm'),grid on, axis([0 60 100 500])

Pulse_dur_256_2=Pulse_dur_tripler+(Dis_coeff_256.*BW.*(29.82.*10.^-6).*10.^-12) % Pulse duration at 256nm [sec]%

d. Code 4 (Windowfouling.m)

% Matlab Code 4
% Models the systematic error due to window unbalance
% Section 7.2: Figure 15
clc
clear all
close all

j=100; % increment for plots
i=19; % increment for E must be 9, 19, 29...
k=1;
m=1;

I_1_b=50e-3; % Initial irradiance of 256nm beam before optics [W/m^2]%
I_4_b=20e-3; %Final irradiance of 256nm beam after optics [W/m^2]

T_max=1; %max total transmission [%]
T_min=0; %min total transmission [%]
T_total=[T_min:(T_max-T_min)/(j-1):T_max]; %matrix for total transmission

E=[-1:2/(i-1):1]; %unbalance parameter;
%if E=0; equally balanced windows%
%if E=1; complete window unbalance%

while m<=i
    while k<=j
        if E(1,m)==0
            Eq=m;
        end
        T_1(k,m)=T_total(1,k)^((1+E(1,m))./2);
        T_2(k,m)=T_total(1,k)^((1-E(1,m))./2);
        k=k+1;
    end
    k=1;
    m=m+1;
end

I_2_b=I_1_b.*T_1; %Initial irradiance [W/m^2]
I_3_b=I_4_b./T_2; %Irradiance over the absorption length [W/m^2]
Beta=(1./I_3_b-1./I_2_b); %measured Beta [m^2/W]

wavelength=256e-9; %[m]%
sigma=4.0e-43; %[m^4] Xe absorption cross-section%
h=6.626e-34; %Planck's constant [J-s]%
c=3e8; %[m/s] speed of light in a vacuum%
v=1/wavelength; %laser wavenumber [m^-1]%
G=1; %second-order coherence function of the photon fields%
Freq=80e6; %Repetition rate of laser [Hz]%
Delta_v_laser=15258.8; %FWHM of laser line, assumes Fourier-limited pulse [m^-1]%
Delta_v_Xe=0*1(100); %[m^-1] absorption line FWHM (Neglected)%
C=sqrt((4*log(2))/(pi*(2*Delta_v_laser^2+Delta_v_Xe^2))); %convolution assuming gaussian [m]%
M2=1; %M^2 value
w_0=4.8; %diffraction limited beam waist, calc from ZEMAX [um]%
w_real=w_0*sqrt(M2)*10^-6; %real beam waist [m]%
L=2*pi*w_real^2/(wavelength.*M2); %absorption length [m]%

n=Beta.*(h*c^2*v)./(2*sigma*G*C.*L); %xenon number density [atoms/m^2]%
m=1;
while m<=i
    n_eq(:,m)=n(:,Eq);
    m=m+1;
end
N_Xe_ratio=n./n_eq;

m=1;
figure
subplot(2,1,1)
hold
while m<=Eq
    plot(T_total(1,2:j)',N_Xe_ratio(2:j,m)),...
    ylabel('N_X_e \epsilon = \epsilon / N_X_e \epsilon = 0'),...
    title('Systematic Error due to Window Unbalance'),...
    text(.1,.99,'\uparrow \epsilon = 0')
    text(.85,.92,'\leftarrow \epsilon = 1')
    axis([T_total(1,2) T_total(1,j) .9 1]);
    m=m+1;
end
m=1;
subplot(2,1,2)
hold
while m<=Eq
    plot(T_total(1,2:j)',N_Xe_ratio(2:j,m)),...
    ylabel('N_X_e \epsilon = \epsilon / N_X_e \epsilon = 0'),...
    xlabel('Total Transmission'),...
    axis([T_total(1,2) T_total(1,j) 0 1]);
    m=m+1;
end

e. Code 5 (Prism.m)

>>Matlab code 5%
%Results for first generation optical setup
% appendix II: Figure 24

clec
clear all
close all

i=1;
j=50; %increment in pressure for plots
%j=10 % used for calc error

x=1.0; %assume pure xenon with 1 atm of N2
P_max=135/14.7; % max total pressure [atm]
P_low=39/14.7; %min total pressure [atm]
P=[P_low:(P_max-P_low)/(j-1):P_max]; %matrix for total pressure [atm]
P%=[39 50 60 70 80 90 100 110 120 135]./14.7; %use for calculating mean error betwn theoretical and measured results
k=1.38e-23; % Boltzmann constant [J/K]%
T=298; %[K]%
n=x.*P.*1.01325e5./(k.*T); %number density of Xe [atoms/m^3]%

wavelength=256e-9; %[m]%
sigma=4e-43; %[m^4] Xe absorption cross-section%
h=6.626e-34; %Planck's constant [J-s]%
c=3e8; %[m/s] speed of light in a vacuum%
v=1/wavelength; % laser wavenumber [m\(^{-1}\)]
G=1; % second-order coherence function of photon field
Freq=80e6; % repetition rate of laser [Hz] 80MHz
Delta_v_laser=15258.8; % FWHM of laser line, assumes Fourier-limited pulse [m\(^{-1}\)]
Delta_v_Xe=0*(100); % [m\(^{-1}\)] absorption line FWHM (Neglected)
C=sqrt((4*log(2))/(pi*(2*Delta_v_laser^2+Delta_v_Xe^2))); % convolution assuming gaussian [m]

Pulse_max=400e-15; % max pulse duration [sec]
Pulse_min=100e-15; % min pulse duration [sec]
Pulse_dur=[Pulse_min:(Pulse_max-Pulse_min)/(j-1):Pulse_max]'; % matrix for pulse dur [sec]

Power=50*10^-3; % Initial average power [W]
M2=3.639; % M^2 value
w_0=4.8; % diffraction limited beam waist, calc from ZEMAX [um]
w_real=w_0*sqrt(M2)*10^-6; % real beam waist [m]
L=2*pi*w_real^2/(wavelength.*M2); % absorption length [m]
A_c=pi*w_real^2 % Cross-sectional area of beam [m^2]

M2_2=29.0931; % Measured M^2 value for results
w_real_2=w_0*sqrt(M2_2)*10^-6; % real beam waist [m]
A_c_2=pi*w_real_2^2 % Cross-sectional area of beam [m^2]

I_0=(Power/Freq)./(Pulse_dur.*A_c); % initial irradiance [W/m^2] for M^2=3.639
I_0_M=(Power/Freq)./(Pulse_dur.*A_c_2); % initial irradiance [W/m^2] for M^2=29.03

% creates matrix for multiplication
while i<=j
    I_0_2(1:j,i)=I_0(1:j,1);
    I_0_M_2(1:j,i)=I_0_M(1:j,1);
    i=i+1;
end

L_0=(h*c^2*v)./((2*sigma*G*C).*(I_0*n)); % [m] length at 50% initial irradiance for M^2=3.639
L_0_2=(h*c^2*v)./((2*sigma*G*C).*(I_0_M*n)); % [m] length at 50% initial irradiance for M^2=29.03

I_L=I_0_2./(1+L./L_0); % irradiance over absorption length [W/m^2] for M^2=3.639
I_L_2=I_0_M_2./(1+L./L_0_2); % irradiance over absorption length [W/m^2] for M^2=29.03

absorbance=log(I_0_2./I_L); % 2-photon absorbance of xenon for M^2=3.639
absorbance_2=log(I_0_M_2./I_L_2); % 2-photon absorbance of xenon for M^2=29.03

% results from prism test
abs_prism=[0.00403985 0.006006419 0.006008276 0.007192405 0.007588668
0.007985402 0.009174057 0.010758843 0.011558434 0.015145937]; %2-photon
absorbance%
p_prism=[39 50 60 70 80 90 100 110 120 135]./14.7; %total pressure
[atm]%
n_prism=(p_prism.*1.01325e5./(k.*298)); %xenon number density
[atoms/m^3]%

% error_mean=mean(absorbance(j,:)/abs_prism);

%Figure compares between the measured and theoretical two-photon
absorbance
%for the prism setup
figure
plot(n,absorbance(j,:),'k',n,absorbance_2(j,:)',':k',n_prism,abs_prism
,'ob'),...
xlabel('
fontsize[12]Xe Number Density
[Atoms/m^3]'),ylabel('
fontsize[12]2-photon Absorbance'),...
legend('
fontsize[12]Theoretical
M^2=3.639','
fontsize[12]Theoretical
M^2=29.09','
fontsize[12]Measured'),...
title('
Absorbance'),...
axis([n_prism(1,1) n_prism(1,10) 0 absorbance(j,j)]);

f. Code 6 (ArgonN2.m)

%Matlab Code 6
%Signal strength for three-photon transition in Argon, Section 11.4
%Signal strength for two-photon transition in N2, Section 11.4

clear all
clc
x=1.0; %mole fraction
P=100/14.7; % total gas pressure [atm]
k=1.38e-23; % Boltzmann constant [J/K]%
T=298; %Temperature [K]%
n=x*P*1.01325e5/(k*T); %initial number density [atoms/m^3]%
wavelength=314.5e-9; %Argon three-photon transition wavelength [m]%
h=6.626e-34; %Planck's constant [J-s]%
c=3e8; %[m/s] speed of light in a vacuum%
G=1; %Second-order coherence function of the photon field%
Freq=80e6; %Repetition rate of laser [Hz]%
Delta_w_laser=10100*c*2*pi; %FWHM of laser line [rad/s]%

M2=1; %M^2 value, factor relating the Gaussian beam to a real beam
w_0=4.8; %diffraction limited beam waist, calc from ZEMAX [um]%
M^2 value, factor relating the Gaussian beam to a real beam
w_real=w_0*sqrt(M2)*10^-6; %real beam waist based on M^2 value [m]%
z=2*pi*w_real^2/(wavelength.*M2); %Rayleigh range (approx absorption
length)
Power = 10; % average power of the laser [W]
Pulse_dur = 180e-15; % pulse duration of the laser [s]
I_0 = (Power/Freq)./(Pulse_dur.*A_c); % initial intensity [W/m^2]

% Signal strength for three-photon transition of Argon
G_w = 1/Delta_w_laser*sqrt(2*log(2)/pi); % convolution of the laser line profile [rad/s]%
sigma = 3.27e-92*G_w; %[s-m^6] Argon absorption cross-section%
Gama_Ar = 3*n*sigma*G*wavelength^2/(h^2*c^2);
I_Ar = sqrt(1/(Gama_Ar*z+1/I_0^2)); % irradiance over the absorption length [W/m^2]
absorbance_Ar = log(I_0/I_Ar); % three-photon absorbance of Argon
Beta_Ar = 1/I_Ar-1/I_0; % Beta for Argon

% Signal strength for two-photon transition of N2
Gama_N2 = 2e-41; % [m^4/W]
I_N2 = 1/(Gama_N2*n*z+1/I_0); % irradiance over the absorption length [W/m^2]
absorbance_N2 = log(I_0/I_N2); % two-photon absorbance of N2
Beta_N2 = 1/I_N2-1/I_0; % Beta for N2