Tunable Diode Laser Absorption Spectroscopy of Hypersonic Flows

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Abstract

Over the past 30 years, researchers have developed tunable diode laser absorption spectroscopy (TDLAS) systems that use low power, continuous wave, tunable diode lasers to measure absorption. Designing a TDLAS system requires a wide-ranging knowledge base covering lasers, optics, electronics, physics and chemistry. Before TDLAS systems become commonplace on vehicles, they need to be well understood, modelled and optimised. Having some design tips and guidelines would help to reduce the time; cost and risk when developing a custom TDLAS system. This paper seeks to summarise best practices and reduce the time required to develop a working TDLAS system.

Introduction

Optical imaging of hypersonic flows provides qualitative information on the fluid flow and can provide quantitative data on density, temperature, pressure, velocity and species concentration. Over the past 30 years, researchers have developed tunable diode laser absorption spectroscopy (TDLAS) systems that use low power, continuous wave, tunable diode lasers to measure absorption. These lasers are more compact than others and can measure absorption at multiple wavelengths allowing researchers to simultaneously measure multiple characteristics of the flow. Eventually, compact TDLAS systems may be fitted to hypersonic vehicles to monitor combustor and isolator parameters and maximise the thrust and stall margin of scramjets.

Designing a TDLAS system requires a wide-ranging knowledge base covering lasers, optics, electronics, physics and chemistry. This means that either a team of experts must design the system from scratch, or a previously developed system is used, or the system is designed and built based on best-practice from the published literature. All three approaches have benefits and limitations. In the first case, the system will probably work first time but the cost and time required to acquire the expertise may be prohibitive. The second approach has the least risk but may limit the experimental freedom of the researchers. The third approach has the highest risk and may require several revisions to account for unexpected problems in getting the system to work.

Before TDLAS systems become commonplace on vehicles, they need to be well understood, modelled and optimised. To achieve this, they should become common in university post-graduate and under-graduate research. Students will either need to use the second or third design approaches. Having some design tips and guidelines would help to reduce the time; cost and risk in students developing bespoke TDLAS systems. This paper seeks to summarise best practices and reduce the time that under-graduates and post-graduates must spend to develop a working TDLAS system.

Theory

When a laser is directed through a test gas, the gas absorbs the radiation in accordance with the Beer-Lambert relation. The Beer-Lambert relation[1] relates the transmitted intensity of a monochromatic laser beam I_ν to the initial intensity I_0:

$$I_\nu = I_0 \exp\left(-k_\nu l\right) \quad (1)$$

where k_ν is the absorption coefficient at wavenumber ν and l is the optical path length. The absorption coefficient is a function of the absorption line strength, $S(T)$, and shape of the line profile, $g(\nu)$, and the number density, N_i of the molecular species, i. The relation is

$$k_\nu = S(T) g(\nu) N_i \quad (2)$$

The line profile depends on temperature and pressure. Pressure changes the shape of the line whilst temperature changes both line strength and shape. If there are overlapping spectral lines, the absorption coefficient is the sum of the influences from each line. If a test gas is irradiated at a frequency near an absorption peak for the target species, the ratio of the transmitted to the initial intensity can be used to calculate the absorption coefficient of the gas.

Line strength, $S(T)$, relative to that at a reference temperature, T_0, is

$$\frac{S(T)}{S(T_0)} = \frac{Q(T)}{Q(T_0)} \frac{\exp(-c_2 E'/T)}{\exp(-c_2 E'/T_0)} \frac{[1-\exp(-c_2\nu_0/T)]}{[1-\exp(-c_2\nu_0/T_0)]} \quad (3)$$

where Q is the total internal partition function calculated by interpolation of data from the HITRAN (high-resolution transmission molecular absorption) database[2], E' is the lower state energy of the transition, ν_0 is the frequency of the transition and c_2 is the second radiation constant, $c_2 = hc/k$, where h is Planck's constant, c is the speed of light[1], and k is Boltzmann's constant.

The gas temperature T may be calculated from the ratio of line strengths of two lines of different E'' and similar ν_0 [1]

$$R(T) = \frac{S_1(T_0)}{S_2(T_0)} \exp\left[-c_2\left(E_1' - E_2'\right)\left(\frac{1}{T} - \frac{1}{T_0}\right)\right] \quad (4)$$

At low pressures, the line profile is Gaussian whilst it has a Lorentzian form at high pressures. Between the two extremes, the line shape may be approximated by a Voigt profile.[1] The Gaussian line shape function is

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[1] In cms^{-1} if the frequency is given in cm^{-1}

$$\Phi\left(\nu^{(j)} - \nu\right) = \frac{\sqrt{\ln 2}}{\sqrt{\pi}\gamma_D^{(j)}} \exp\left\{-\ln 2\left(\frac{\nu^{(j)} - \nu}{\gamma_D^{(j)}}\right)^2\right\} \quad (5)$$

where $\Phi\left(\nu^{(j)} - \nu\right)$ is the intensity of the jth spectral line (that has a centre wavenumber of $\nu^{(j)}$) at a wavenumber of ν and $\gamma_D^{(j)}$ is the Doppler half-width at half maximum of the line which is, in turn, given by:

$$\gamma_D^{(j)} = \frac{\nu^{(j)}}{c}\sqrt{\frac{2\ln 2 kT}{m}} \quad (6)$$

where m is the molecular mass of the test gas. The Doppler width is independent of pressure. At high pressures, the line shape may exhibit pressure broadening resulting in a Lorentzian profile described by

$$\Phi\left(\nu^{(j)} - \nu\right) = \frac{1}{\pi}\frac{\gamma_L^{(j)}}{\left(\nu^{(j)} - \nu\right)^2 + \gamma_L^{(j)2}} \quad (7)$$

where $\gamma_L^{(j)}$ is the air-broadened half-width at half maximum (HWHM$_a$). The Lorentzian halfwidth is a linear function of pressure and varies with temperature as a power relationship

$$\gamma_L^{(j)}\left(T, p\right) = \gamma_L^{(j)}\left(T_0, p_0\right)\frac{p}{p_0}\left(\frac{T_0}{T}\right)^{r(j)} \quad (8)$$

where $\gamma_L^{(j)}\left(T, p\right)$ is the Lorentz half-width of the jth feature at temperature T and pressure p, $r(j)$ is the temperature ratio power for a collision half-width of the jth line, $T_0 = 296$ K and $p_0 = 101.325$ kPa. The Voigt line shape is a convolution of the Doppler and Lorentzian contours and is defined as [1]

$$V_\Phi\left(\nu^{(j)} - \nu\right) = \frac{k_0^{(j)}y^{(j)}}{\pi}\int_{-\infty}^{\infty}\frac{e^{-t^2}}{y^{(j)2} + \left(x^{(j)} - t\right)^2}dt \quad (9)$$

where $k_0^{(j)} = \frac{\sqrt{\ln(2)}}{\sqrt{\pi}\gamma_{D^{(j)}}}$, $y^{(j)} = \sqrt{\ln 2}\frac{\gamma_D^{(j)}}{\gamma_L^{(j)}}$, $x^{(j)} = \sqrt{\ln 2}\frac{(\nu - \nu^{(j)})}{\gamma_L^{(j)}}$.

The algorithm provided by Humlíček [3] can be used to approximate the Voigt profile. When using this algorithm, use x as the real component and y as the imaginary component. The symmetrical Voigt profile is the real component of the algorithm's output. Whiting [4] gives the following approximation[1] of the Voigt HWHM

$$\gamma_V = \frac{\gamma_L}{2} + \sqrt{\left(\frac{\gamma_L^2}{4} + \gamma_D^2\right)} \quad (10)$$

If the laser wavelength is ramped across the feature, the method is called direct absorption spectroscopy. First harmonic detection is defined as having the laser wavelength modulated sinusoidally at low frequency (below about 10kHz) across the feature. Second harmonic detection (or wavelength modulated spectroscopy) is used when a second higher frequency sinusoidal modulation is applied to the low frequency first harmonic

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[1]Accurate to about one percent

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modulation. Direct absorption spectroscopy is easiest to simulate, implement and understand but the output can be difficult to interpret due to noise and changes in the laser diode power as the current changes.

This theoretical derivation of the gas number density is complicated practically by uncertainties in the gas temperature, absorption by species other than the target, chemical reactions in the flow, atmospheric absorption and other sources of noise in the detection electronics.

Application of Theory

Consider a system that uses direct absorption to detect water vapour features in the region near 1392.5335nm ($7181.15578 \text{cm}^{-1}$). Table 1 lists the features at and near the target wavelength. The data includes the wavenumber of the centreline (ν), the centreline wavelength (λ), the intensity (S), the air-broadened half-width at half maximum (HWHM$_a$) of the lineshape, the self-broadened HWHM (HWHM$_s$), the energy of the lower transition level (E') and the temperature coefficient (r). The major feature for this experiment was the strong absorption peak at $7181.15578 \text{cm}^{-1}$ (1392.533nm).

In this application, the aim is to measure the temperature and pressure of a water vapour flow at various conditions. Griffiths [1] recommends calculating the temperature using equation 4 provided that the lines are close to each other. The important parameters are the line strengths and lower state energies for the two selected lines. Line strength ratio is a weak function of temperature making this method suitable when temperature changes over a wide range such as from 300K to 1700K.

When temperature varies over a small range, the feature with the largest absolute rate of change of intensity with respect to temperature has the best temperature sensitivity. For the features in table 1, the line at $7181.15578 \text{cm}^{-1}$ is best for measuring temperatures below about 600K. At these temperatures, that line's temperature sensitivity is four times the next best line. The disadvantage of using a single line to estimate temperature is that the line shape depends on temperature at low pressures and on pressure also at higher temperatures.

Consider measuring the flow properties of water vapour at 2.5kPa and 250K using the feature at $7181.15578 \text{cm}^{-1}$. At this low pressure, the Gaussian profile can be used with a HWHM of $9.58 \times 10^{-3} \text{cm}^{-1}$ as calculated using equation 6. For a flow of pure water vapour, the HWHM$_s$ should be used to calculate the Lorentzian HWHM. From equation 8, γ_L is 0.01375cm^{-1}. This is 1.436 times the Doppler HWHM at the same temperature. The Voigt HWHM is 0.01867cm^{-1} using equation 10. The line intensity at this temperature is $2.033 \times 10^{-20} \text{cm}^{-1}/(\text{molecule} \times \text{cm}^{-2})$ using equation 3 and the total internal partition function of 135.713088.[2] The line shapes and intensities for the other features can be calculated similarly resulting in a plot of absorbance. To calculate the temperature from direct absorption readings, estimate the temperature, generate an absorbance plot and fit the plot to the recorded data. Iterate this process until an acceptable fit is achieved. The pressure cannot be determined using the Gaussian profile but it can be estimated from the Voigt profile.

If the pressure increases to 25kPa, the Gaussian profile is no longer suitable and a Voigt profile should be used. From equation 8, γ_L is 0.13754cm^{-1}. This is 14.36 times the Doppler HWHM at the same temperature. The ten-fold increase in pressure has caused γ_L to increase ten-fold as expected from equation 8. The pressure increase has caused γ_V to increase to 0.1382cm^{-1}.

ν	λ	S	γ_a	γ_s	E_{low}	r
(cm^{-1})	(nm)	(cmmol^{-1})	(cm^{-1})	(cm^{-1})	(cm^{-1})	
7179.75201	1392.8058	1.802×10^{-22}	0.0456	0.265	1216.1945	0.61
7180.39972	1392.6801	5.608×10^{-22}	0.098	0.500	225.8384	0.69
7181.15578	1392.5335	1.505×10^{-20}	0.1008	0.488	136.7617	0.71
7182.20911	1392.32927	1.541×10^{-21}	0.107	0.375	42.3717	0.76
7182.94962	1392.1857	3.752×10^{-21}	0.097	0.51	142.2785	0.71
7185.596571	1391.6729	1.995×10^{-22}	0.0342	0.371	1045.0583	0.62
7185.596909	1391.6728	5.984×10^{-22}	0.0421	0.195	1045.0577	0.62

Table 1. Strong spectral features near the target (Source: [2])

Holding the pressure constant at 25kPa and increasing the flow temperature to 350K causes the line intensity to change, the Doppler width to increase and the Lorentzian width to increase. The influence of the Doppler broadening will again be small compared to pressure broadening but is larger than at the lower temperature. Under these conditions, $\gamma_D = 0.0113\mathrm{cm}^{-1}$, $\gamma_L = 0.1083\mathrm{cm}^{-1}$ and $\gamma_V = 0.1095\mathrm{cm}^{-1}$ for the feature at $7181.15578\mathrm{cm}^{-1}$.

As the HWHM increases, more energy is in the wings of the profile, lowering the peak absorbance. Thus, the feature wings are important when determining the pressure. The relationship between pressure, temperature and HWHM is complicated and depends on the properties for each feature. These examples have given a flavour of the relationships between the parameters. as a further complication, the measurements of water vapour may have been performed in the presence of another gas such as air, N_2 or CO_2. In each case, the HWHM due to the carrier gas changes along with the temperature coefficient. If taking measurements with a different carrier gas, ensure that you use the correct values for that gas or another similar gas.

Getting Started

Hobbs [5] recommends that, when designing an optical instrument, you start by writing down the things that you know including the basic physics. From this, you can determine the most important operating impacts. Next, devise the measurement principle and estimate the photon budget, the detection area required and laser power required. After this, make a rough optical design choosing numerical apertures, wavelengths, distances and optical components. The next step is to devise a detection and post-processing strategy, make a detailed photon budget and check whether the system will work. If not, revise your design. Proceeding with a design that will not work will waste your time, effort and limited funds.

Before starting to design the system, the science requirements must be decided. These drive all following system design decisions. Specify the experimental conditions in terms of pressure, temperature, species concentration, carrier gases and path length. Using the equations above, estimate absorbances and determine the laser diode power and photo-detector responsivity needed to achieve an acceptable signal-to-noise ratio (SNR) for the system.

Hobbs [5] recommends that TDLAS signals should be processed electronically where possible as this provides the best SNR. As may be inferred from the foregoing, having enough power and a low SNR are the two most important aspects in developing a functional TDLAS system. Where possible, components should be chosen to maximise the intensity of the detection beam whilst minimising optical and electronic noise.

Wavelength Selection

A target species must be in sufficient concentration and must absorb or emit radiation at a suitable wavelength to be a candidate for TDLAS monitoring. Each target species has numerous absorption or emission peaks depending on the reactions taking place and on the flow temperature and pressure. To determine the optimal measurement wavelength, consult a modelling program such as HITRAN[2], Specair[6] or HITEMP[7]. HITRAN is an excellent source for atmospheric spectra. HITEMP is analogous to the HITRAN database but encompasses many more bands and transitions than HITRAN for the absorbers H_2O, CO_2, CO, NO, and OH. Specair is less rigorously verified than the other two databases but it provides a user-friendly interface whilst modelling the absolute intensity of spectral radiation emitted by gases and plasmas.

Wavelength choice also depends on the availability of laser diodes, photodetectors and optics. Choosing a strong absorption peak may be irrelevant if there are no commercially available laser diodes or photodetectors at that wavelength. This is particularly relevant for detecting water vapour as many of the cheapest and most readily available laser diodes have been developed for electronics or telecommunications and tend to avoid wavelengths where water vapour absorbs strongly. In the end, the wavelength choice may be a compromise between the best theoretical choice and one where optical components are commercially available.

Laser Source

Various suppliers (including Thorlabs, MPC, SRS, Newport) provide laser diode controllers off-the-shelf. When choosing a controller consider the maximum power required, thermal and current stabilities, the number of diodes to be separately controlled and the interfaces required to other systems.

Laser diodes mounts provide the thermal and electrical interfaces between the diode and the controller. Mounts have thermoelectric controllers (TECs) to stabilise the diode temperature and centre wavelength. The diode current may be modulated to enable harmonic detection. Some mounts can inject the radio frequency (RF) modulation directly into the laser diode. If this facility is used, ensure that the laser diode is capable of RF modulation. To detect harmonic output data, the controller should be able to modulate the diode current using a saw-tooth waveform at 10kHz or more. This waveform should in turn be modulated using a sinusoidal signal at frequencies of over 1MHz for short time-frame experiments in shock tunnels. The sinusoidally modulated saw-tooth waveform is required to detect the harmonic output data.

Laser Diodes

Being the heart of a TDLAS system, the laser diode is extremely

important and should be carefully selected. Important parameters are the type of modulation (that is, pulsed or continuous wave (CW)), wavelength, power, spectral width, current tuning coefficient, temperature tuning coefficient and the beam divergence. Generally, CW diodes are used for TDLAS because they can be modulated over the test time. Laser diode powers vary from approximately 1mW to more than 2W. The power level should be selected to provide an adequate signal-noise ratio (SNR) for the test system.

Spectral width of the laser diode should be as small as possible but is typically 100kHz-300MHz. Always ask the supplier if the diode can be used for spectroscopy. The current tuning coefficient is between 0.1-2nmA^{-1} and the temperature tuning coefficient is typically 1nmK^{-1}. Laser diodes come in various configurations of pin numbers, packages, and diode layouts. The most common types are three-pin 5.6mm and 9mm TO packages or butterfly packages. The mount must be compatible with the diode configuration. It is important to check this prior to ordering a mount or diode as it may be a unpleasant surprise if they are not compatible after waiting up to three months for delivery. Beam divergence may be up to 30° horizontally and 60° vertically. This should be immediately collimated via an aspheric lens and, if necessary, an anamorphic prism pair to circularise the beam. Inserting a Faraday isolator as soon as possible in the optics train will reduce feedback from downstream elements.

The laser diode output wavelength and power will typically change with the diode temperature and current. Manufacturers can provide calibration charts or curves for diodes but these can incur some cost. Even so, the diode calibration may change with time so you should know how to calibrate your diodes to ensure that you are actually measuring what you think you are measuring. To calibrate your diode, identify three or four strong spectral features within the tuning range of the diode. If you plan to use a high current with your diode, set the diode temperature and turn the laser on well before the test. As the diode heats, the TEC will take time to stabilise the diode temperature. This, in turn, changes the power output and wavelength of the laser. Carefully, methodically and repeatably change the diode current in small increments across the tuning range and record the absorption features. When changing the diode current, ensure that the TEC temperature stabilises before taking measurements. Use the feature data to fit calibration curves to the diode parameters. It is very important to have accurate and precise wavelength calibration as it provides the basis for all future measurements.

If the laser wavelength of a diode that emits into free space coincides with a strong atmospheric absorption band for water vapour or CO_2, the optics train should be enclosed in an inert gas environment. Optical interference due to unintentional etalons can severely impact on measurements. Where possible, angled fibre-optic connectors and wedged windows should be used and the target area windows should have anti-reflective coatings.

Signal Detection

Having successfully transmitted the modulated laser beam(s) into the target area, we now need to detect the output and process the data to determine the flow temperature, relative line strengths and species concentration. The design guidelines for the receiver optics reflect those for the transmitter with some additional requirements. The optical train should be enclosed in an inert atmosphere if the laser wavelength is susceptible at room temperature to absorption by atmospheric species such as water vapour. During shock or expansion tunnel tests, consid-

erable radiation is emitted from the test area. Any unwanted sources of radiation, such as background emissions from the test area, should be filtered out prior to detection.

The refractive index, density and other flow parameters in the test area may change significantly during test. The optical train should accommodate these variations in the beam path. The test area windows, the filter and the photodetector should be tuned to reduce etalons in the optical path. Some windows that are transparent at visible wavelengths absorb strongly and non-linearly in the near-infrared. Measure and, if possible, calibrate the absorbance due to the windows across the wavelength range of interest.

The photodetector should be placed as close as practicable to the test area. Its active area should be larger than the laser beam cross-section to ensure as much of the signal as possible is detected. If the photodetector area is smaller than the laser beam cross-section, additional optics may be needed to focus the beam onto the active area. Ensure that the power incident on the photodiode does not exceed its damage threshold. The photodetector should have a fast rise-time (say 60ns) to ensure that rapid changes in intensity are detected. This is particularly important for pulsed hypersonic test facilities where the test time may be as short as 50μs. The responsivity of a photodiode is typically 0.2-1.2AW^{-1}.

Conclusions

This paper has provided design guidelines that should assist post- and under-graduate students to design and develop TDLAS systems that work first-time without the students having to spend excessive time in researching literature. The system described is simple but the principles can be extended to more complex systems.

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