LASER DIAGNOSTICS FOR
SIMULTANEOUS VAPOR AND DROPLET
MEASUREMENT IN SPRAYS

By
Lin Ma

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High Temperature Gasdynamics Laboratory
Department of Mechanical Engineering
Stanford University
Stanford, California 94305
Abstract

Rapid and reliable measurements of vapor and droplets in sprays are desirable in many scientific and industrial applications. This work describes the development of such measurement methods based on wavelength-multiplexed laser absorption and extinction, with several practical applications.

Diode-laser sensors based on spectrally-resolved absorption were first developed for the measurements of hydrocarbon fuel vapor, including mole fraction and temperature of the vapor. Utilizing the overtone band of the C-H vibrational band, these sensors enabled sensitive and reliable monitoring of ethylene vapor with temporal resolution better than 0.2 ms. The reliability and rapid time response of these sensors have promoted much success in several measurement campaigns, including monitoring of the fueling process and active engine control in pulse detonation engines.

The vapor measurement method was then extended to a more general sensing concept, namely Wavelength-Multiplexed Laser Extinction (WMLE), for the simultaneous measurement of multiple parameters in sprays with Sauter mean diameter ranging from sub-micron to about 10 µm. Based on the measurement of extinction at multiplexed-wavelengths ranging from 0.6 to 10 µm, the WMLE scheme was developed to provide simultaneous characterization of both vapor and droplets in water sprays, including vapor concentration, vapor temperature, droplet size distribution, and droplet volume fraction. This WMLE diagnostic was first demonstrated in cold-flow spray systems, then in an aerosol shock tube to simultaneously measure water aerosol and vapor properties during interactions between shock waves and the aerosols. Extension of the sensing technique to other spray systems beyond water is analyzed.

These successful demonstrations provide strong support for the further development and expanded applications of such sensing techniques.
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Chapter 1

Introduction

1.1 Background and Motivation

Rapid and reliable measurements of vapor and droplets simultaneously in sprays are desirable in many scientific and industrial applications. The study of vaporization and combustion of liquid fuel sprays is a typical example.

Numerous laser-based techniques have been developed for the separate characterization of either vapor or droplets. These vapor sensing techniques include laser Rayleigh scattering [1], spontaneous Raman spectroscopy [1], laser-induced fluorescence (LIF) [1, 2], spectrally resolved absorption spectroscopy [3], and others. Droplet measurement techniques include the measurement of Fraunhofer diffraction of laser radiation by droplets [4], laser phase Doppler anemometry [5], and laser extinction [6]. Each technique has its own advantages and disadvantages and is useful for a certain domain of applications.

However, these techniques are generally not transferable to the simultaneous monitoring of both vapor and droplet properties. Some vapor sensing techniques based on elastic scattering such as the Rayleigh scattering technique or LIF can be extended to measure both vapor and droplets. But, usually the scattering from droplets greatly exceeds that from the vapor, therefore jeopardizing or even prohibiting an accurate vapor measurement. Similar reasoning precludes the direct combination of one vapor and one droplet measurement technique discussed above for simultaneous vapor and droplet measurements. Furthermore, different techniques usually have
different temporal and spatial resolution, and some of the techniques may require wide optical access, so that practical implementation becomes difficult.

Despite these challenges, a few laser-based diagnostic techniques have been attempted for simultaneous characterization of vapor and droplets. Laser Induced Exciplex Fluorescence (LIEF) developed by Melton [7] allowed two-dimensional imaging of vapor and droplets simultaneously under limited conditions. Besides implementation difficulties, however, the LIEF signal is difficult to quantify, and quenching from oxygen limits most of the LIEF applications to nitrogen environments [1]. For more quantitative and realistic applications, Chraplyvy and Tishkoff [8, 9] proposed and applied a method based on a two-wavelength laser extinction strategy to measure vapor concentration and droplet volume fraction simultaneously in an evaporating fuel spray.

Of the methods investigated thus far, techniques based on laser extinction are especially attractive owing to their relative simplicity in implementation and data interpretation, capability to provide continuous measurement with fast time response, very limited requirement of optical access, and species specificity if wavelengths are well-chosen. These techniques are based on the transmission measurement of narrow-linewidth laser radiation by the spray, which is described by the following relationship:

\[
T(\lambda) = \frac{I_r}{I_0} = \exp[-\tau_{\text{vapor}}(\lambda)] \cdot \exp[-\tau_{\text{drops}}(\lambda)]
\]  
(1.1)

where:

- \( T(\lambda) \) = the optical transmittance at wavelength \( \lambda \)
- \( I_0 \) and \( I_r \) = the incident beam intensity and the transmitted beam intensity
- \( \tau_{\text{vapor}}(\lambda) \) = the vapor absorbance
- \( \tau_{\text{drops}}(\lambda) \) = the droplet extinction
The vapor absorbance and droplet extinction in Equation (1.1) depend on properties of the vapor (e.g., concentration and temperature) and droplets (e.g., size distribution and volume fraction) respectively. Therefore, by measuring the optical transmission at well-selected wavelengths, properties of vapor and droplets can be inferred.

In this work, a diagnostic technique utilizing laser extinction at multiplexed wavelengths has been developed and demonstrated for the simultaneous vapor and droplet measurement in water sprays. Extension of this diagnostic technique to hydrocarbon sprays is also considered and analyzed. Compared with previous work, this wavelength-multiplexed laser extinction (WMLE) diagnostic considers the following two extensions for enhanced sensing capability in sprays. The first extension considered incorporates a vapor sensing technique modified from spectrally-resolved absorption spectroscopy to significantly improve the detection limit of vapor sensing. The second extension considered includes laser sources at a larger number of wavelengths in a wide spectral range to expand sensing capability of the diagnostic to more quantities besides vapor concentration and droplet volume fraction, such as vapor temperature and size distribution of the droplets. This thesis will introduce the theories of the diagnostic technique, describe the implementation of the theories, and demonstrate the diagnostic in different applications.

1.2 Organization of Thesis

This thesis first describes the development of laser diagnostics for the separate characterization of vapor and droplets, then the development of WMLE schemes for the simultaneous characterization of vapor and droplets.

More specifically, the present chapter provides the motivation and outline for the thesis and a brief review of past work. Chapter 2 introduces the principles of spectrally-resolved absorption spectroscopy for vapor measurement, followed by the development and applications of
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an ethylene vapor sensor. Chapter 3 is devoted to a theoretical discussion of droplet measurement by laser extinction. Chapter 4 extends the discussions in Chapter 2 and 3 to the simultaneous measurement of vapor and droplets in sprays. Chapters 3 and 4 illustrate the design of the WMLE scheme through the example of water sprays, with consideration and analysis of the extension of the WMLE technique to other spray systems beyond water. Chapter 5 describes the development of WMLE schemes based on the theories developed in Chapters 3 and 4, with experimental demonstrations of such scheme in water sprays. Chapter 6 summarizes the thesis and suggests some future directions of this research.

References


Chapter 2

Vapor Measurement Using Spectrally Resolved Absorption Spectroscopy

2.1 Introduction

Among the diagnostic methods developed for vapor sensing, techniques based on direct absorption are especially attractive owing to the relative simplicity of implementation and data interpretation. These techniques are based on the absorption of narrow-linewidth laser radiation by the probed medium, which is described by the Beer-Lambert relationship:

\[ \frac{I_t(\nu)}{I_0(\nu)} = \exp[-k(\nu) \cdot L] \]  

(2.1)

where \( I_0(\nu) \) and \( I_t(\nu) \) are the incident and transmitted laser intensities at frequency \( \nu \), \( k(\nu) \) the spectral absorption coefficient, taken as uniform across the region probed, and \( L \) is the pathlength. The product \( k(\nu)L \), which equals \(-\ln[I_t(\nu)/I_0(\nu)]\), is known as the spectral absorbance. In the case of spectrally isolated individual transitions, the spectral absorption coefficient, \( k(\nu) \), can be expressed as:

\[ k(\nu) = \frac{P_{\text{total}}}{X} \cdot S(T, \nu_0) \cdot \phi(\nu-\nu_0) \]  

(2.2)

where \( P_{\text{total}} \) is the total static pressure of the mixture, \( X \) the mole fraction of the absorbing species along the pathlength, \( S(T, \nu_0) \) the line strength of the transition centered at frequency \( \nu_0 \), and \( \phi(\nu-\nu_0) \) the line-shape function. At a given temperature, it can be shown that the mole fraction of the absorbing species is proportional to the integrated spectral absorbance of the probe beam,
obtained by modulating the frequency of the probe beam across one or more absorption features [1], as shown in Equation (2.3)

\[ \int_{v_1}^{v_2} \ln \frac{I_d(v)}{I_0(v)} dv = P_{\text{total}} \cdot X \cdot \sum_i S(T, v_i) \cdot L \]  

(2.3)

where \( v_1 \) and \( v_2 \) defines the frequency scan range of the probing beam, and \( S(T, v_i) \) represents the strength of the \( i^{th} \) transition of the absorbing species between \( v_1 \) and \( v_2 \). Therefore, if the total line strengths of all the transitions within the scan range are known from measurement or simulation, the mole fraction of the absorbing species can be inferred by measuring the integrated spectral absorbance, provided that the pressure and pathlength are known. Extension of this technique to address nonuniformity along the probe beam will be discussed in Chapter 6.

This chapter describes the development of laser diagnostics using spectrally resolved absorption spectroscopy for vapor sensing. As an example, the development of a \( \text{C}_2\text{H}_4 \) (ethylene) sensor is detailed. Then we present the applications of this sensor to characterize the fueling process in two practical engines. In the first application, this sensor was employed to characterize the fuel stoichiometry in a research pulse detonation engine (PDE) at Stanford University. Based on such characterizations, an active control scheme based on this sensor was demonstrated to improve the fueling process in this engine. In the second application, this sensor was used to diagnose the fuel filling process in a multi-cycle PDE at the General Electric (GE) Global Research Center at Niskayuna, NY.

### 2.2 Ethylene Vapor Sensor

#### 2.2.1 Motivation and Background

Ethylene (\( \text{C}_2\text{H}_4 \)) is a hydrocarbon vapor commonly encountered in many applications. In the study of PDE physics and engine operation, gaseous \( \text{C}_2\text{H}_4 \) has been used extensively as a fuel...
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[2], owing to its easy detonability and convenient delivery to laboratory engines. The wide use of
C$_2$H$_4$ has motivated the development of a robust C$_2$H$_4$ sensor.

Previously, a C$_2$H$_4$ sensor based on 3.39 µm HeNe laser (fixed-wavelength) absorption
was developed and used in propulsion and shock tube applications [3,4,5]. However, the fixed-
wave length of the laser sources at 3.39 µm limits the applications of these sensors. For example,
the accuracy of this sensor can be compromised by the hostile measurement environment in PDEs
[6] where noise sources such as beamsteering (i.e., reorientation of the probing laser beam due to
gradients in the refractive index normal to the optical path) and broadband absorbers and
scatterers (such as liquid fuel droplets and soot) are commonly encountered. In this section, we
describe the development of a C$_2$H$_4$ sensor based on tunable diode-laser aimed at overcoming the
deficiencies of the 3.39 µm technique. Note that with the continuing development of new laser
sources (such as tunable laser sources near 2 and 3.39 µm), other C$_2$H$_4$ sensing strategies have
become available since the work described here.

2.2.2 Description of the Ethylene Sensor

The absorption spectrum of gaseous ethylene C$_2$H$_4$ is rich with a variety of distinct
features. Past effort on the study and measurement of these spectral features laid the foundation
for the detection of C$_2$H$_4$ using diode-laser absorption spectroscopy. Survey spectra of C$_2$H$_4$
measured by Brock et al. indicate that three strong absorption bands occur in the spectra range
from 2800 to 15000 cm$^{-1}$ [7]. The strongest absorption feature occurs in the C-H fundamental
band between 2900 and 3300 cm$^{-1}$, which can be simulated by the HITRAN database [8]. Figure
2.1 shows the fundamental band of C$_2$H$_4$ simulated by HITRAN 2004. As shown in Figure 2.1, a
distinct absorption peak occurs near 2990 cm$^{-1}$. The other two strong absorption features occur in
the combination bands of C-H between 4000 and 5000 cm$^{-1}$ and in the first overtone of C-H
between 5600 and 6400 cm$^{-1}$, respectively. Measurements of the combination and overtone bands
with high spectral resolution reveal that the $\nu_5 + \nu_{12}$ band (between 4514 and 4565 cm$^{-1}$) and the $\nu_5 + \nu_9$ band (between 6150 and 6197 cm$^{-1}$) provide the strongest absorption among the combination bands [9]. Although the band intensity of the $\nu_5 + \nu_{12}$ band is about two times that of the $\nu_5 + \nu_9$ band, lack of a convenient laser source around the $\nu_5 + \nu_{12}$ band hindered the use of this absorption feature at the time when this work was conducted. Therefore, the fundamental and $\nu_5 + \nu_9$ combination band are of primary interest for the development of C$_2$H$_4$ detection techniques based on absorption spectroscopy.

The HeNe laser at 3.39 $\mu$m (2950 cm$^{-1}$) provides a convenient light source to exploit the strong absorption in the fundamental band. Sensors based on this wavelength also have the advantage that they can be readily used to detect other hydrocarbon vapors. These potential advantages have motivated several past measurements of the absorption coefficients of various hydrocarbon vapors [10] and their temperature and pressure dependences [11, 12] and applications of C$_2$H$_4$ sensors based on the 3.39 $\mu$m wavelength in combustion environments [13-15]. Unfortunately, absorption at 3.39 $\mu$m by C$_2$H$_4$ is significantly weaker than other hydrocarbon vapors [11, 12]. For example, absorption of 3.39 $\mu$m HeNe radiation by C$_2$H$_4$ is more than 30 times weaker than that of propane (C$_3$H$_8$) and butane (C$_4$H$_{10}$), and about 100 times weaker than heptane (C$_7$H$_{16}$) vapor. Weak absorption impairs the detection limit of C$_2$H$_4$ at this wavelength, and interference absorption from other trace hydrocarbon contaminants becomes a potential problem. As a fixed-wavelength light source, the 3.39 $\mu$m HeNe laser cannot be wavelength-tuned to access other stronger absorption features nearby in the fundamental band, as shown in the inset of Figure 2.1.

Tunable lasers offer the capability to select the optimum absorption feature and avoid interference absorption. Furthermore, sensors based on tunable lasers also have other important advantages over sensors based on a fixed-wavelength strategy. One such advantage is the ability of the sensor to better discriminate noises commonly encountered in combustion applications,
such as beamsteering, broadband absorbers and scatterers (e.g., soot particles and liquid fuel droplets) [16, 17]. Another advantage is that tunable lasers have the potential to enable simultaneous monitoring of multiple parameters of the vapor (such as vapor temperature, concentration, and pressure) [18]. These considerations motivated our development of a C$_2$H$_4$ sensor based on the absorption feature between 6150 and 6197 cm$^{-1}$ ($\sim$ 1.62 $\mu$m) in the $\nu_5 + \nu_9$ combination band, where tunable diode lasers are readily available from the telecommunications industry. These semiconductor lasers are also attractive owing to their very low noise, compatibility with optical fibers, and relatively low cost.

The absorption spectra of C$_2$H$_4$ in the $\nu_5 + \nu_9$ band were measured by temperature-tuning a diode laser (SEI SLT5411-DA-S460) as shown in Figure 2.2. Among all the features in the combination bands, the Q-branch of the $\nu_5 + \nu_9$ band is attractive for an optimum C$_2$H$_4$ sensing strategy in that 1) the Q-branch offers the strongest absorption and 2) the absorption strength of the Q-branch decays more slowly than other features. Sample measurements of part of the Q-branch (between 6151.5 and 6143.4 cm$^{-1}$) are shown in Figure 2.3 and Figure 2.4 for different temperatures and pressures of a mixture of C$_2$H$_4$ and N$_2$. These measurements are recorded by single scans of the diode laser in 0.2 ms. Note the variation in the detailed shape of this feature with temperature and pressure. Therefore, the spectral information recorded by this sensor has the potential to enable simultaneous measurement of multiple parameters of C$_2$H$_4$ fuel. To speed the spectral fitting needed to determine temperature, we selected the ratio of two peaks illustrated in Figure 2.3 for this purpose. The ratio at different temperatures is shown in Figure 2.5, fit to a Boltzmann distribution model. This simple scheme has proven very effective to monitor C$_2$H$_4$ temperature in PDE flows. In Figure 2.6, the integrated absorbance, as defined in the following equation, of the feature in Figure 2.4 is plotted:

$$\text{Integrated absorbance} = \int_{v_1}^{v_2} \text{Absorbance} \cdot dv$$ (2.4)

where
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\[ \nu_1, \nu_2 = \text{the lower and higher frequency bound of the absorption feature scanned} \]
\[ = 6143.35 \text{ and } 6151.54 \text{ cm}^{-1}, \text{ respectively} \]

Absorbance = the product of spectral absorption coefficient and pathlength as defined in equation (2.1)

The linear relationship between the integrated absorbance and pressure enables this C\textsubscript{2}H\textsubscript{4} sensing strategy to be insensitive to pressure. From the measurements shown in Figure 2.6, the total line strength of all the transitions in this absorption feature at 296.8 K is determined to be 0.420 cm\textsuperscript{-2}/atm (from 6143.35 to 6151.54 cm\textsuperscript{-1}). Therefore, at a temperature of 296.8 K, the mole fraction of C\textsubscript{2}H\textsubscript{4} can be inferred from the measured integrated absorbance by the following relationship

\[ X_{C2H4} = \frac{\text{Integrated absorbance (cm}^{-1})}{0.420 \text{ cm}^{-2}/\text{atm} \cdot P_{\text{total}} \text{(atm)} \cdot L \text{(cm)}} \]  (2.5)

Similar relationship can be developed to infer the mole fraction of C\textsubscript{2}H\textsubscript{4} at other temperatures. Determination of the total line strength used in Equation (2.5) at other temperatures requires absorption measurements of the Q-branch at the corresponding temperatures, such as those measurements shown in Figure 2.3.

2.3 Application in Stanford PDE

2.3.1 Experimental Arrangement

Figure 2.7 provides a schematic of the experimental set-up in the Stanford PDE. The fuel (C\textsubscript{2}H\textsubscript{4}) and oxidizer (O\textsubscript{2}) are metered with flow orifices and premixed before injection into the PDE. The firing sequence begins with simultaneous opening of the fuel and oxidizer valves, subsequent filling of the PDE, closing of the valves, electric spark ignition, and a delay time interval (before repeating the cycle) for the burnt gases to exhaust.
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The C_2H_4 sensor is applied to the head and tail ends of the engine simultaneously. Information obtained at these two measurement stations is sufficient to characterize and control the C_2H_4 distribution in the engine when C_2H_4/O_2 input flow rates are well-behaved. However, more measurement stations can readily be incorporated by adding additional beam splitters and detectors. The distributed feedback (DFB) laser’s current tuning range is extended to obtain an 8 cm^{-1} scanning range, sufficient to record C_2H_4’s Q-branch spectral feature near 6150 cm^{-1}. Transmitted DFB laser beams are monitored by InGaAs detectors. Detector voltages are digitized using 12-bit resolution for quantitative absorption measurements, or detected with a lock-in amplifier synchronized with the diode laser wavelength sweep rate for control experiments. When the lock-in signal exceeds the set point, the PDE firing sequence is initiated.

2.3.2 Characterization of the Fueling Process

Characterization of the fuel distribution is of fundamental importance for the development of PDEs. To validate PDE models and computations, uniform fuel charges are desired and the quantitative fuel/oxidizer stoichiometry is needed. For the purpose of improving engine performance, fuel charges must be intelligently tailored to avoid wasted fuel, to achieve the desired specific impulse, and to create an easily detonable fuel distribution. In the Stanford PDE, we demonstrated quantitative fuel concentration measurements and active fuel-loading control.

The C_2H_4 concentration data recorded at the tail measurement station in the Stanford PDE is shown in Figure 2.8 for two C_2H_4/O_2 engine fills, one with and one without ignition. Such data contain a wealth of important information. For instance, the fuel concentration does not rise instantaneously, owing to the finite valve opening time and to diffusion processes that occur as the fuel flows through the PDE tube. Obviously we could use such measurements to select valves and tailor their control signals to optimize performance. The transient signal corresponds to plug flow of a specific fuel concentration. If a uniform fuel distribution is desired, then the fuel must...
be loaded at least to the tail end of the engine, which may result in wasted fuel (as in the case shown in Figure 2.8). In a practical engine application, wasted fuel could be avoided by adjusting the timing of valve operation and ignition to ensure that no fuel has exited the tail end of the PDE prior to arrival of the detonation wave.

### 2.3.3 Active Control of the Fueling Process

To optimize fuel consumption and to control specific impulse, we designed an active control system based on the C₂H₄ sensor to control the valve operation and ignition timing.

Figure 2.7 illustrates the experimental arrangement in the Stanford PDE to actively control valve operation and ignition timing. We modulated the wavelength over the rapidly varying feature in the C₂H₄ spectrum shown in Figure 2.2 near 6150 cm⁻¹. For rapid time response of the control system, analog detection, rather than digital data acquisition, is used to analyze the detector signal. A lock-in amplifier is employed to process the diode laser absorption signal synchronously with the diode laser wavelength modulation. The laser absorption is low when C₂H₄ concentration is low, therefore the output of the lock-in amplifier is low; similarly, high C₂H₄ concentration produces high absorption and lock-in output signal. When the output of the lock-in amplifier reaches a set decision level (it corresponds to the C₂H₄ concentration reaching a desired value at the tail end), we initiate the firing sequence: the valves are closed, and the igniter is triggered to fire the fuel/oxidizer charge. If the decision level is properly chosen, valves are closed and the engine is fired when the PDE tube is just fully filled with C₂H₄/O₂ mixture. Hence, fuel usage is optimized and constant impulse can be obtained with minimal fuel waste for each cycle.

We demonstrated this control system in the PDE for the case of finite-volume supply tanks. As the supply tanks exhaust their contents, the fuel/oxidizer flow rates decline with the supply tank pressure. If the PDE charging time is set to a constant fill-time schedule, the impulse
(time integral of force on tube head end, measured with a pressure gauge) decreases for each fill. Figure 2.9 compares the fuel fill duration and impulse per PDE cycle for a scheduled case and the actively controlled case. Figure 2.9a shows how the fuel fill duration must be increased to accommodate the decreasing fuel supply pressure. As a result, the impulse is nearly constant for each pulse, as shown in Figure 2.9b, because of the full tube fill ensured by the active control. In contrast, the impulse from a scheduled fill decreases on each PDE cycle. By changing the tail end measurement station to another location, a similar control scheme could be used to realize partial fuel fills in the PDE; non-uniform spatial fuel distribution may be achieved with non-constant valve flow-rates. All these forms of fuel-fill tailoring are best accomplished with a quantitative fuel sensor. See Chapter 6 for discussions on the development of schemes to provide qualitative fuel control information.

As noted, the broad wavelength-scanning technique employed renders the sensor relatively immune to noise imposed by the harsh PDE environment, such as beamsteering or window effects. The low-noise level immediately after the detonation wave arrival, shown in Fig. 6, demonstrates this immunity, which is critical whenever the equivalence ratio of a PDE charge must be known or controlled.

2.4 Application in GE PDE

Prior to the implementation of this C\textsubscript{2}H\textsubscript{4} sensor, combustion events in the PDE facility at the GE Global Research Center were classified qualitatively as “detonation / no detonation”. The identification of the optimum operating fuel/air ratio of the PDE facility was attempted based on the sound produced by the combustion event, with clear “pops” being the indicator of a “good” detonation. This method led to some success in identifying optimum operating conditions, but clearly the need existed for a more qualitative measurement of fuel-air stoichiometry, and further understanding of the valve behavior and ignition timing of the PDE facility.
This need led to the application of this diode laser C$_2$H$_4$ sensor in the GE PDE during the period of Dec. 15 to 19, 2003. This sensor showed great performance as it was easy to align and robust during shipment and repeated engine operation. The sensor was applied to both the head and tail ends of the benchmark PDE tube operating on C$_2$H$_4$/air at 10 Hz for both cold flow and hot fired experiments. More than 50 experiments were performed to cover the whole range of operation conditions of interest to GE PDE research.

2.4.1 Experimental Arrangement

The PDE tube configuration with the C$_2$H$_4$ sensor applied is schematically shown in Figure 2.10. It consists of a 5.0 cm diameter, 1.27 m long stainless steel tube. This configuration is valveless on the air side, and fuel flow is modulated with a high-speed solenoid valve. Fuel and air are mixed at the head end of the tube and a spark plug is located just downstream of the fuel-air mixing element. There are obstacles in the tube to promote transition to detonation. The 1.62 µm diode laser sensor was used separately at both the head and tail end measurement locations in order to characterize the fuelling process of the PDE tube. On the pitch side of the sensor, as shown in Figure 2.11, the laser source is coupled into a fiber, and the fiber is held in a specially-machined mount. The mount is rigidly attached to the engine via threaded bolts, so that it moves in phase with any mechanical vibrations induced in the tube structure. On the detection side, an integrating sphere and an InGaAs detector are mounted on an L-shaped bracket, and the bracket is also rigidly attached to the engine. This optical engineering of the sensor has proved to be easy to align, robust during shipment and repeated experiments, and resistant to noise caused by beamsteering, emission, and mechanical vibration. Note that bandpass filters can also be incorporated on the detection side to help removing emission. In these experiments we found noises caused by emission is insignificant compared with the laser intensity except during the
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period (about 0.2 ms) when the detonation wave passes the probe beam. Therefore, no filter was used in the experiments.

The 1.62 μm diode laser is current-modulated at 5 kHz during experiments to scan the Q-branch absorption features shown in Figure 2.2 from 6143.35 to 6151.54 cm⁻¹. The transmitted laser beam is monitored by an InGaAs detector, then converted into spectral absorbance and finally integrated over the modulation frequency to infer the C₂H₄ concentration based on Equation (2.4).

2.4.2 Example Measurement

This sensor was applied to both the head and tail ends of the PDE tube operating on C₂H₄/air at 10 Hz for both cold-flow and hot-fired experiments. More than 50 experiments were performed to cover a wide range of operating conditions. These measurements provide a wealth of valuable information to characterize the fuel/air stoichiometry, valve timing, and ignition timing of the PDE tube. For example, Figure 2.12 shows results of the sensor applied at the tail end of the engine operating at 10 Hz with steady air mass flow of 0.042 kg/s. In this set of experiments, the ethylene supply pressure is increased from 100 to 180 psi in steps of 20 psi, while the steady air mass flow rate is held constant and the fuel/air flow is maintained at chocked flow condition. As shown in Figure 2.12, the sensor clearly resolves the increase in equivalence ratio \( \phi \) due to the increase in fuel supply pressure. It is also evident that, as the supply pressure in the 0.25 inch fuel supply line drops, the valve cannot supply as much fuel and the equivalence ratio drops quite markedly within one second of a given test. In the experiment with a fuel supply pressure of 160 psi, the fuel-air stoichiometry begins at roughly \( \phi = 1.1 \) but quickly drops to about \( \phi = 0.6 \) as the pressure in the fuel supply line drops. This results in an unsteady and dynamically-changing fill from cycle to cycle during repeated engine operation, namely that the first few cycles have a markedly different stoichiometry than those that will follow, making comparisons
difficult for multi-cycle experiments. The sensor was able to diagnose this non-ideal behavior, and as a result steps have been taken to improve the fuel delivery system. The measurements in Figure 2.12 also show that ignition occurs about 60 ms after the arrival of the fuel-air mixture at the tail measurement location. This delay indicates that a certain amount of the fuel-air mixture has been loaded beyond the engine at the time of ignition and does not contribute to the generation of thrust. If one desires to fully fill the engine while maximizing the fuel usage, the ignition timing should be adjusted to minimize this delay and this sensor is of obvious utility for this task.

Figure 2.13 shows the fuel-air stoichiometry measured by the laser sensor at different fuel supply pressures and a fixed air flow rate of 0.042 kg/s. As we have seen above in Figure 2.12, the value of $\phi$ drops throughout the one second of data acquired as the supply pressure in the 0.25 inch line depletes. For consistency, the initial value of $\phi$ at the time of the first spark has been chosen as the sensor-determined value of $\phi$. The sensor results are compared to predictions from simple one-dimensional bulk flow calculations, and show good agreement at low ethylene supply pressure. Thus, as a first-order approximation, bulk flow can be used to determine an optimum setpoint of fuel supply pressure given a fixed air flow rate. From Figure 2.13 we see that a fuel supply pressure of about 130 psi at an air flow rate of 0.042 kg/s results in an equivalence ratio of unity.

To further validate the sensor’s results and show the utility of the sensor to verify the operation of the engine, the measurements from the diode-laser were compared with the flame emergence time measured previously by GE. GE performed a series of experiments to determine the time required for the combustion wave front to emerge from the PDE tube as a function of equivalence ratio. The emergence time is defined as the time it takes for the combustion wave to exit the tail end of the PDE tube, relative to when the spark is ignited (time zero). These emergence time measurements were made in a clear polycarbonate plastic tube using a high-
speed camera which directly visualizes the luminosity from the spark discharge as well as the chemiluminescent emission from the hot wave front traveling the length of the tube [19]. A counter is started when the spark discharges, and is stopped when the combustion wave reaches the tail end of the tube. The time difference from the counter determines the emergence time. Figure 2.14 shows the comparison between flame emergence time measurements and diode laser sensor measurements. The flame emergence time is measured with the PDE operating at an air flow rate of 0.042 kg/s and with a range of ethylene supply pressures. These experiments show a minimum emergence time at a particular fuel supply pressure (130 psi) at this air mass flow rate. These engine operation conditions were then repeated with the diode laser fuel sensor applied to the tail end monitoring the equivalence ratio for each experiment. Figure 2.14 shows a clear correlation between the flame emergence time and the equivalence ratio, namely that the minimum in the flame emergence time curve occurs at roughly $\phi = 1.0$, as determined by the laser sensor. This independent measurement serves as further validation of the sensor’s results and shows the utility of the sensor to both verify the operating point of previous experiments, and to predict the ideal operating point of future experiments.

2.5 Summary

In conclusion, a diode-laser absorption sensor has been developed and used to monitor the fuel filling process in two PDEs, one operating on oxygen-C$_2$H$_4$ and one air-breathing PDE for cycle frequencies up to 10 Hz. In both applications, this sensor provided quantitative measurements of fuel concentration with high temporal resolution to characterize the fuel-air stoichiometry, the fuel delivery system, and the valve behavior. Active control of valve and ignition timing under changing injection conditions has also been successfully demonstrated in the Stanford PDE using this sensor. The control scheme optimized fuel consumption and
achieved constant impulse for each cycle. During these applications, the sensor demonstrated robust and reliable operation. The success achieved in these applications provides further support for the use of diode laser sensors in practical propulsion systems.

References:


Figure 2.1. Absorption spectrum of C\textsubscript{2}H\textsubscript{4} near 3.39\textmu m simulated by HITRAN 2000. Simulation conditions: T=300 K, P=1 atm, 25% C\textsubscript{2}H\textsubscript{4} and 75% O\textsubscript{2}.

Figure 2.2. Absorption spectrum of C\textsubscript{2}H\textsubscript{4} near 1.62 \textmu m for a mixture of 25% C\textsubscript{2}H\textsubscript{4} and 75% N\textsubscript{2} at 296 K and 1 atm. Measured by temperature-tuning a DFB diode laser.
Figure 2.3. Q-branch of $\text{C}_2\text{H}_4$ near 1.62$\mu$m at different temperatures. Measured in a static cell with 20 cm absorption path length, pure $\text{C}_2\text{H}_4$ and 1 atm pressure.

Figure 2.4. Q-branch absorption spectrum of $\text{C}_2\text{H}_4$ near 1.62 $\mu$m measured by diode laser sensor in 0.2 ms for each scan.
Figure 2.5. Ratio of the two peaks indicated in Fig. 2 to monitor temperature at 1 atm.

Figure 2.6. Integrated line strength of the Q-branch of $\text{C}_2\text{H}_4$ spectra shown in Figure 2.2.
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Figure 2.7. Schematic of C$_2$H$_4$ sensor applied to Stanford PDE for monitoring and control.

Figure 2.8. Record of fuel concentration at tail-end measurement station with and without ignition.
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Figure 2.9. Active control experiments based on diode laser fuel sensor to realize a full tube fill in Stanford PDE. (a) comparison of fuel fill duration, and (b) comparison of impulse per cycle between fixed duration case and active control case.

Figure 2.10. Experimental arrangement of C₂H₄ measurement at the PDE facility at General Electric (GE) Global Research Center at Niskayuna, NY.
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Figure 2.11. Schematic of the C₂H₄ sensor designed for PDE measurement at GE.

Figure 2.12. Example C₂H₄ fuel history measurements at tail measurement location. Air flow rate = 0.042 kg/s, Repetition rate = 10 Hz, C₂H₄ supply pressure varied from 80 to 180 psi.
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Figure 2.13. Comparison of laser measurement of $\phi$ to simplified bulk flow rate analysis.

Figure 2.14. Comparison of flame emergence time with equivalence ratio determined by the laser sensor. The minimum time of emergence is seen to occur near $\phi = 1.0$ as expected.
Chapter 3

Droplet Measurement Using Laser Extinction

3.1 Introduction

Currently there is a strong interest in using liquid hydrocarbon fuels in pulse detonation engines (PDEs), owing to their high energy density, stability, and safety in transportation and storage. Fuel aerosols with small characteristic mean diameters (below 10 µm typically) are of particular interest in these applications [1-3] because of the improved PDE efficiency and easy detonability when such small fuel droplets are used. This research interest created the need for the development of non-intrusive optical sensors and motivates this work.

Appendix A provides a survey of different methods for aerosol measurement. Among all the optical methods for the measurement of aerosol size distribution functions (SDFs), techniques based on Mie extinction are especially well-suited for this application, owing to their relative simplicity in implementation, capability to provide continuous measurement with high temporal resolution, and very limited requirement of optical access. However, most past work on the measurement of SDFs by the extinction method relied on light sources from a relatively narrow wavelength range (usually in the spectral range from the near-UV to the near-IR) [4-8], resulting in two constraints on the performance of SDF measurements. First, the amount of information that can be inferred about the SDFs from extinction measurements is very limited if these measurements are performed at wavelengths within a narrow spectral range. Twomey et al. [6] found that extinction measurements based on wavelengths from the near-UV to the near-IR allow
only one or two independent inferences about the SDFs for droplets with diameter less than 10 µm. Second, extinction measurements in a narrow spectral range limit the sensitivity of the SDF measurements and the applicable range of the extinction method.

The continuing development of laser technologies facilitates the consideration of incorporating wavelengths in a wider spectral range in the extinction method [9, 10]. Here we extend the extinction method to a general wavelength-multiplexed laser extinction (WMLE) concept by incorporating an arbitrary number of wavelengths in a wide spectral range and investigated the potential advantages brought about by such extension to overcome the above constraints. An example WMLE scheme utilizing wavelengths ranging from 0.25 to 10 µm was investigated for the SDF measurements of water aerosols to illustrate these advantages. This work has shown that inference of multiple parameters about the SDFs is allowed by extinction measurements performed at well-selected wavelengths in this extended spectral range. This scheme was found to enable determination of a variety of SDFs having Sauter mean diameter ranging from sub-micron to about 10 µm with enhanced sensitivity compared to extinction schemes based on a more limited wavelength range. The sensitivity analysis performed here also provides useful guidance for the selection of wavelengths in the WMLE scheme to achieve optimum SDF measurements. Compared with previous work, this work has the following two unique aspects. First, this work provides a systematic way of evaluating and selecting wavelengths from a wide spectral range (0.25 to 10 µm) for droplet sizing using laser extinction. The sensitivity of different wavelengths in the measurement of droplet size and distribution is analyzed for different spray systems and distribution function. Second, this WMLE concept shows promise to enable a relatively simple and stable inversion of the extinction data to SDFs.

3.2 Measurement Concept

The governing equations for the measurement of SDFs by Mie extinction are as follows:
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\[ \tau_i = -\ln\left(\frac{I_i}{I_0}\right) = \frac{\pi}{4} C_n L \int_0^\infty Q(\pi D / \lambda_i, m) f(D) D^2 dD \]  

(3.1)

where:

\( \tau_i \) = the extinction by the aerosols at wavelength \( \lambda_i \)

\( I_0 \) and \( I_i \) = the intensity of incident and transmitted light at wavelength \( \lambda_i \)

\( C_n \) = the number density of the aerosols

\( L \) = the pathlength

\( Q(\pi D / \lambda_i, m) \) = the extinction coefficient of an aerosol with diameter \( D \) at wavelength \( \lambda_i \)

\( m \) = the complex refractive index of the aerosols at wavelength \( \lambda_i \)

\( f(D) \) = the aerosol size distribution function defined such that \( \int_0^\infty f(D) dD = 1 \), and

\( f(D) dD \) represents the probability that an aerosol has diameter between \( D \) and \( D + dD \).

Note that Equation (3.1) assumes uniformity of the aerosol properties along the line of light and only single scattering is considered here. Also note that in practice, the extinction signal detected will inevitably contain a certain amount of contribution from spurious scattering due to the finite detection angle/area. See Appendix B for discussions on the applicable range of the single scattering assumption and the evaluation of the interference scattering in the extinction signal.

The determination of the SDFs then reduces to a solution of Equation (3.1) for \( f(D) \) based on extinction measurements performed at selected wavelengths. However, Equation (3.1) is ill-conditioned and the development of a stable algorithm to invert the extinction measurements to SDFs has long been a subject of research effort [4, 9-11]. These algorithms can be divided into two categories. In the first category, no \textit{a priori} information about the sought SDF is available; and in the second category, \textit{some} \textit{a priori} information is available (for example, the SDF can be described by an empirical distribution function). When the sought SDF can be described by an known function, \( f(D) \), Equation (3.1) can be modified to the following equations:
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$$R_{ij} = \frac{\tau_i}{\tau_j} = \frac{\bar{Q}(\lambda_i, D_{32})}{\bar{Q}(\lambda_j, D_{32})}$$  \hspace{1cm} (3.2)

where $R_{ij}$ is the ratio between measured extinction at wavelength $\lambda_i$ and $\lambda_j$, $\bar{Q}$ and $D_{32}$ are the mean extinction coefficient and Sauter mean diameter defined in Equation (3.3) and (3.4) respectively:

$$\bar{Q}(\lambda_i, D_{32}) = \int_0^\infty \frac{Q(\pi D / \lambda_i, m)f(D)D^2 dD}{\int_0^\infty f(D)D^3 dD}$$  \hspace{1cm} (3.3)

$$D_{32} = \int_0^\infty \frac{f(D)D^3 dD}{\int_0^\infty f(D)D^2 dD}$$  \hspace{1cm} (3.4)

The log-normal function, as defined in Equation (3.5), is one of the most commonly used SDFs and will be used as an example distribution function to introduce the measurement concept

$$f(D) = \frac{1}{\sqrt{2\pi D \ln \sigma}} \exp[-\frac{1}{2(\ln \sigma)^2} \cdot (\ln D - \bar{D})^2]$$  \hspace{1cm} (3.5)

where $\sigma$ characterizes the distribution width and $\bar{D}$ the mean size of the distribution. Application of the WMLE to other distribution functions will be discussed in Section 3.6. Figure 3.1 shows four log-normal distribution functions having the same $D_{32}$ with $\sigma$ varied over a relatively wide range. It can be seen from Figure 3.1 that the diameter spans a decade at a $\sigma$ of 1.6. In Figure 3.2, the mean extinction coefficients ($\bar{Q}$'s) of water aerosols at a temperature of 22 °C following log-normal distributions with the $\sigma$'s shown in Figure 3.1 are calculated and compared to the extinction coefficients of a mono-dispersed distribution ($\sigma$=1). Refractive indices used in the calculations are from [12, 13] and are listed in the caption of Figure 3.2. Figure 3.2 illustrates that $\bar{Q}$ for a log-normal distribution with a $\sigma$ of 1.1 has an almost identical profile as that of a mono-

\* Due to symmetry, $\lambda_i > \lambda_j$ is assumed in this work.
dispersed distribution, especially at long wavelengths such as 10 µm. At relatively short wavelengths (e.g., 0.5 and 4 µm), the difference is in the fine ripple structures. The similarity of these profiles indicates that an attempt to distinguish log-normal distributions with σ around 1.1 or less from a mono-dispersed distribution by the extinction method would require measurements at very high accuracy to resolve the fine structures in the extinction coefficient curves. Therefore, here we limit our consideration for σ to the range of 1.2 to 1.6, and the extension to wider ranges of σ will be discussed later in this paper.

Different methods with various levels of complexity have been created to solve Equation (3.2) [4, 8, 14] to yield stable solution of SDFs within certain ranges of mean diameter and distribution width. Observations made in Figure 3.2 inspire a relatively simple method to determine SDFs based on extinction measurements. As shown in Figure 3.2, \( \bar{Q} \) for \( \lambda=10 \) µm exhibits insensitivity to the distribution width for \( D_{32} \) in the range up to 11 µm, and so does \( \bar{Q} \) at \( \lambda=0.5 \) µm for \( D_{32} \) greater than about 3 µm. As a result, the ratio defined in Equation (3.2) between these two wavelengths will not be sensitive to distribution width for \( D_{32} \) ranging from 3 to 11 µm. Thus, information about \( D_{32} \) in this range can be extracted from extinction measurements at these two wavelengths in the absence of detailed knowledge about the distribution width. This observation can be generalized to other combinations of wavelengths by analyzing the definition of \( \bar{Q} \) in Equation (3.3). It can be shown that \( \bar{Q} \) defined in Equation (3.3) is independent of f(D) when Q is a linear function. This analysis provides a mathematical explanation for the insensitivity of \( \bar{Q} \) at 10 µm for \( D_{32} \) up to 11 µm, because Q at 10 µm behaves almost linearly in this diameter range. It is obvious that \( \bar{Q} \) defined in Equation (3.3) is also independent of f(D) when Q is a constant. This explains the insensitivity of \( \bar{Q} \) at 0.5 µm for \( D_{32} \) greater than 3 µm, because Q at 0.5 µm is asymptotic to a constant in this range. Therefore, for any given two wavelengths, when Q at each wavelength meets one of these two conditions (Q can
be approximated by a linear function or a constant) over a common $D_{32}$ range, the ratio defined in Equation (3.2) between these two wavelengths is insensitive to the distribution width and can be used to determine $D_{32}$ without knowledge about the distribution width in this $D_{32}$ range.

After $D_{32}$ is measured, information about the distribution width ($\sigma$) can be obtained by extinction measurements at wavelengths which exhibit sensitive dependence on distribution width. For example, $\bar{Q}$ at 4 $\mu$m exhibits sensitivity to distribution width for $4 < D_{32} < 10$ $\mu$m but $\bar{Q}$ at 0.5 $\mu$m does not, therefore the ratio of extinction measurements between these two wavelengths should be sensitive to $\sigma$ and can be used to measure the distribution width in this diameter range. Measurements of distribution width in other $D_{32}$ ranges can be obtained by incorporation of other wavelengths.

Finally, $D_{32}$ of a log-normal distribution is related to the parameters of the distribution by the following equation,

$$\ln \bar{D} = \ln D_{32} - \frac{5}{2}(\ln \sigma)^2$$

(3.6)

Therefore $\bar{D}$ is determined after $D_{32}$ and $\sigma$ are determined, and so is the distribution function.

In the following two sections, we discuss the application of the above method to measure $D_{32}$ and distribution width of log-normal distributions in greater detail, and evaluate the sensitivity of such measurements.

### 3.3 $D_{32}$ Measurement

As discussed in Section 3.2, $D_{32}$ can be measured by a combination of two wavelengths in the range over which the ratio of $\bar{Q}$ at these wavelengths displays insensitivity to distribution width ($\sigma$). In Figure 3.3, the ratio of $\bar{Q}$ shown in Figure 3.2 at 0.5 and 10 $\mu$m are calculated for three distribution widths ranging from 1.2 to 1.6. As expected, this ratio exhibits overall
insensitivity to $\sigma$ for $D_{32}$ up to about 11 $\mu$m and therefore enables $D_{32}$ measurements in this range. For example, when the ratio of extinction between these two wavelengths is measured to be 0.3, $D_{32}$ can be determined to be about 6.8 $\mu$m ($\pm 0.1$ $\mu$m) without detailed knowledge of $\sigma$. However, closer observation of Figure 3.3, as shown in the inset, reveals that ambiguity exists in the determination of $D_{32}$ due to the lack of detailed knowledge of $\sigma$, especially in the $D_{32}$ range where $R$ shows more significant variations with $\sigma$ (such as in the range of $D_{32}>12$ $\mu$m). Therefore, we need to quantify the ambiguity in the $D_{32}$ measurement to specify the applicable range of a given pair of wavelengths and to estimate the measurement uncertainty. As illustrated in the inset of Figure 3.3, the largest ambiguity occurs when the actual distribution has a $\sigma$ of 1.2 (the lower limit of the $\sigma$ under consideration) while the $R$ calculated at a $\sigma$ of 1.6 (the upper limit of the $\sigma$ under consideration) is used to determine $D_{32}$, or vice versa. Denote the $D_{32}$ determined by the $R$ calculated at the lower limit of the distribution width $D_{32}^1$, and that determined at the upper limit $D_{32}^2$. Then the difference between $D_{32}^1$ and $D_{32}^2$ ($\Delta D_{32}$) as shown in Equation (3.7) represents the largest possible ambiguity in the $D_{32}$ measurement

$$\Delta D_{32} = D_{32}^1 - D_{32}^2$$ \hspace{1cm} (3.7)

The relative ambiguity is usually of more interest in most applications. Therefore, the average of $D_{32}^1$ and $D_{32}^2$ ($D_{32}^{avg}$) is used to approximate $D_{32}$ and the ratio between $\Delta D_{32}$ and $D_{32}^{avg}$ defined in Equation (3.8) is used to quantify the relative ambiguity in $D_{32}$ measurement

$$\frac{\Delta D_{32}}{D_{32}} \approx \frac{\Delta D_{32}}{D_{32}^{avg}} = \left(\frac{D_{32}^1 - D_{32}^2}{D_{32}^1 + D_{32}^2}\right)/2$$ \hspace{1cm} (3.8)

Figure 3.4 shows the calculation of $\Delta D_{32}/D_{32}$ when the extinction measurements at 0.5 and 10 $\mu$m are used to determine $D_{32}$ of water aerosols following log-normal distributions. Figure 3.4 indicates that $D_{32}$ can be determined within 5% in the range from about 3.4 to 11.7 $\mu$m for $\sigma$ in the range of 1.2<$\sigma$<1.6 by these two wavelengths. Obviously, better knowledge of $\sigma$ (i.e.
narrower range of possible $\sigma$) reduces the ambiguity in $D_{32}$ measurement or extends the range over which $D_{32}$ can be determined within fixed level of ambiguity, as demonstrated by the calculation performed for $\sigma$ in the range of 1.2 to 1.4 and 1.4 to 1.6. Note that Figure 3.4 also shows that $\Delta D_{32}/D_{32}$ is minimized at $8 < D_{32} < 10$ $\mu$m, a region where the $\bar{Q}$ curves at 10 $\mu$m corresponding to different distribution widths intersect as shown in Figure 3.2.

Similar methods can be used to determine $D_{32}$ in other ranges. This concept is illustrated through the following example design of a WMLE scheme. Assume the availability of light sources with wavelengths ranging from 0.25 to 10 $\mu$m to determine $D_{32}$ within 5% in the range of $1 < D_{32} < 10$ $\mu$m for water aerosols following log-normal distributions with $1.2 < \sigma < 1.6$ (at the same time, the number of wavelengths required should be minimized). Forty wavelengths uniformly spaced between 0.25 and 10 $\mu$m (the spacing is 0.25 $\mu$m) are considered in this conceptual design. As shown in Figure 3.4, a combination of wavelengths of 0.5 and 10 $\mu$m satisfies all the requirements for $3.4 < D_{32} < 11.7$ $\mu$m. Additional wavelengths need to be selected to measure $D_{32}$ in the range of $1 < D_{32} < 3.4$ $\mu$m. This selection is virtually a trial-and-error process, though the analyses performed in Section 3.2 to predict the behavior of $\bar{Q}$ provide insights into this process and substantially reduce the number of trials. The results of the wavelengths selected are summarized in Figure 3.5, with the combination of wavelengths of 0.5 and 10 $\mu$m to measure $D_{32}$ in the range of $3.4 < D_{32} < 11.7$ $\mu$m, 0.5 and 4 $\mu$m to measure $1.5 < D_{32} < 3.4$ $\mu$m, and a pair of relatively short wavelengths, 0.25 and 1.5 $\mu$m, to measure $0.7 < D_{32} < 1.5$ $\mu$m. A WMLE scheme based on these five wavelengths would satisfy all the requirements of this design. In practice, other criteria may also require consideration. For example, $R$ between the selected wavelengths should behave monotonically on the corresponding $D_{32}$ range to allow unique determination of $D_{32}$, the value of $R$ should be between 0.1 and 10 to ensure optimum signal to noise ratio of the
measurements, and $D_{32}$ should be measured with reasonably high sensitivity ($S_D$) defined in the following equation:

$$S_D = \left| \frac{dR / R}{dD_{32} / D_{32}} \right| \sigma$$  \hspace{1cm} (3.9)

Calculations show that the above WMLE scheme also satisfies these additional criteria with a $S_D$ greater than unity for $1.2 < \sigma < 1.6$.

Hence, in summary, a WMLE scheme composed of five wavelengths, 0.25, 0.5, 1.5, 4, and 10 $\mu$m, will enable sensitive and unique determination of $D_{32}$ in the range of $0.7 < D_{32} < 11.7$ $\mu$m within 5% in the absence of detailed knowledge about $\sigma$ for water aerosols following log-normal distributions with $\sigma$ in the range from 1.2 to 1.6. More wavelengths in and beyond this interval would of course enhance the measurement accuracy, and would accommodate expansions in the ranges of $D_{32}$ and $\sigma$.

### 3.4 Distribution Width Measurement

As mentioned in Section 3.2, after $D_{32}$ is measured, $\sigma$ can be determined by a combination of two wavelengths in the $D_{32}$ range where $R$ between these wavelengths exhibits sensitivity to $\sigma$. For example, Figure 3.2 shows that $\bar{Q}$ at 4 $\mu$m is sensitive to the distribution width for $D_{32}$ in the range of $4 < D_{32} < 10$ $\mu$m while $\bar{Q}$ at 0.5 $\mu$m is not in this $D_{32}$ range. Therefore, $R$ between these two wavelengths should be sensitive to $\sigma$ for $D_{32}$ in the range from 4 to 10 $\mu$m, as confirmed by the calculations shown in Figure 3.6. Figure 3.6 shows the ratios of $\bar{Q}$ between these two wavelengths for water aerosols following log-normal distributions with various distribution widths. According to Figure 3.6, the $R$ curves vary substantially with $\sigma$ for $D_{32}$ in the range of $4 < D_{32} < 10$ (especially in the vicinity of $D_{32} = 7.3$ $\mu$m) and less significantly in other $D_{32}$
ranges (especially in the vicinities of $D_{32}=3.1$ and 10.6 µm, where the R curves corresponding to different distribution widths intersect).

A quantified evaluation of the sensitivity of $R$ to $\sigma$ is necessary before applying these wavelengths to measure distribution widths. Variations of $R$ with $\sigma$ are calculated at selected values of $D_{32}$s mentioned in Figure 3.6 and shown in Figure 3.7 to provide insights into the quantification of the sensitivity for $\sigma$ measurements. As expected, the $R$ curve is considerably more sensitive to $\sigma$ at $D_{32} = 7.3$ µm than at $D_{32} = 10.6$ µm. Moreover, the $R$ curve at $D_{32} = 10.6$ µm does not behave monotonically with $\sigma$, and consequently cannot yield unique determination of $\sigma$ at this $D_{32}$. A quantity ($S_{\sigma}$) defined in Equation (3.10) provides a measure of the sensitivity and helps to recognize this nonmonotonic behavior

$$S_{\sigma} = \left| \frac{dR}{R} \right|_{D_{32}}$$

A large $S_{\sigma}$ implies that a small proportional change in $\sigma$ results in a large proportional change in $R$ at the given $D_{32}$, therefore sensitive determination of $\sigma$ is enabled at this $D_{32}$, and vice versa. If a $S_{\sigma}$ of zero occurs, it typically implies that the corresponding $R$ curve is not monotonic at the given $D_{32}$ over the $\sigma$ range under consideration. As illustrated in Figure 3.7, in the $\sigma$ range from 1.2 to 1.4, $S_{\sigma}$ corresponding to $D_{32}=7.3$ µm is greater than 0.6 while $S_{\sigma}$ corresponding to $D_{32}=10.6$ µm is less than 0.3. Therefore, the ratio of $\bar{Q}$ between these two wavelengths allows more sensitive determination of $\sigma$ at $D_{32}=7.3$ µm than at $D_{32}=10.6$ µm. Moreover, the $S_{\sigma}$ curve at $D_{32}=7.3$ µm has a zero value near $\sigma=1.4$, which implies that the corresponding $R$ curve is not monotonic and cannot provide a unique determination of $\sigma$ at this $D_{32}$. In summary, Figure 3.7 shows that the wavelength combination of 0.5 and 4 µm provides a sensitive and unique determination of $\sigma$ at $D_{32}$ near 7.3 µm, but application of this wavelength combination at $D_{32}$ near 10.6 results in low sensitivity and ambiguity of the $\sigma$ determination.
However, generalization of the above discussion is limited because the $S_\sigma$ defined in 
Equation (3.10) depends on multiple variables ($D_{32}$ and $\sigma$). In order to reduce the number of 
dimensions in the evaluation of $S_\sigma$, we define $S_\sigma^{\text{min}}$ as the minimum of $S_\sigma$ in the range of $\sigma$ under 
consideration at a given $D_{32}$, and use $S_\sigma^{\text{min}}$ to represent the sensitivity of $\sigma$ measurement at this 
$D_{32}$. Clearly, $S_\sigma^{\text{min}}$ is a conservative estimation of the sensitivity for $\sigma$ measurements, though it 
still contains the information to help recognize the nonmonotonic behavior of the $R$ curve. Figure 
3.8 shows the calculation of $S_\sigma^{\text{min}}$ for the wavelength combination of 0.5 and 4 $\mu$m for distribution 
widths ranging from 1.2 to 1.6. As expected from Figure 3.6 and Figure 3.7, this $S_\sigma^{\text{min}}$ curve 
peaks near $D_{32}=7.3$ $\mu$m and has zero values near $D_{32}=3.1$ $\mu$m and $D_{32}=10.6$ $\mu$m. For a preset 
minimum sensitivity requirement, the applicable range of this wavelength for $\sigma$ measurement can 
be decided by the calculation of $S_\sigma^{\text{min}}$ shown in Figure 3.8. For example, if a $S_\sigma^{\text{min}} \geq 0.31$ is 
required, then according to Figure 3.8, the wavelength combination of 0.5 and 4 $\mu$m can be 
applied for $\sigma$ measurements in the range of $0.4 < D_{32} < 1.4$ $\mu$m and $4.7 < D_{32} < 9.5$ $\mu$m. Figure 3.8 
also presents the $S_\sigma^{\text{min}}$ calculations for two other wavelength combinations (10 and 4 $\mu$m, and 1.5 
and 0.25 $\mu$m). According to our calculations, among all the possible combinations between the 
wavelengths selected for $D_{32}$ measurements in the previous section, these two pairs of 
wavelengths (10 and 4 $\mu$m, and 1.5 and 0.25 $\mu$m) provide the optimum $\sigma$ measurements for $D_{32}$ in 
the range from 0.4 to 9.5 $\mu$m for $\sigma$ in the range of $1.2 < \sigma < 1.6$. More specifically, as shown in 
Figure 3.8, the wavelength combination of 1.5 and 0.25 $\mu$m would enable measurements of $\sigma$ for 
$D_{32}$ in the range of $1.8 < D_{32} < 3.9$ $\mu$m, the combination of 10 and 4 $\mu$m for $0.4 < D_{32} < 1.8$ $\mu$m and 
$3.9 < D_{32} < 9.5$ $\mu$m, and the minimum sensitivity for the $\sigma$ measurements is greater than 0.31 for $\sigma$ 
in the range of 1.2 to 1.6. These two wavelength combinations are also examined to confirm that 
they satisfy the requirement of $0.1 < R < 10$. Similar to the discussion of $D_{32}$ measurements,
additional wavelengths are needed if a higher $S_{\sigma}^{\text{min}}$ is required or the ranges of $D_{32}$ and $\sigma$ to be measured are expanded.

As mentioned before, Figure 3.8 also implies that the incorporation of wavelengths from a wide spectral range expands the applicable range of the extinction method and enhances the sensitivity of SDF measurements. For example, in an extinction scheme where only wavelengths in the range of 0.25 to 1.5 $\mu$m are utilized, the measurements of SDFs with $D_{32}$ above about 4 $\mu$m would be difficult, and the limited wavelength range also impairs the sensitivity of the measurements.

Figure 3.9 summarizes all the wavelengths selected in Section 3.3 and 3.4 for the WMLE scheme to measure SDFs of water aerosols following log-normal distributions, the applicable $D_{32}$ ranges for each of the wavelength combinations for optimized SDF measurements, and all the criteria considered in the wavelength selection process. This scheme enables determination of log-normal distribution functions in the range of $0.7 < D_{32} < 9.5$ $\mu$m and $1.2 < \sigma < 1.6$ with good sensitivity ($S_D > 1.0$ and $S_{\sigma}^{\text{min}} > 0.31$) and signal to noise ratio ($0.1 < R < 10$). According to our previous studies, extinction measurements at these selected wavelengths are mutually independent and therefore do not contain experimental redundancy. Moreover, the method used in this scheme to invert extinction measurements to SDFs is inherently stable. Uncertainties in the SDF measurements are derived from the ambiguity in $D_{32}$ measurements ($\Delta D_{32}/D_{32}$), which is bounded, and the bounds can be estimated based on the calculations shown in Figure 3.5. For example, at a $D_{32}$ of 3.5 $\mu$m, as shown in Figure 3.5, application of the wavelength combination of 4 and 0.5 $\mu$m results in a $\Delta D_{32}$ which is -5% of the $D_{32}$ to be measured, and application of the combination of 10 and 0.5 $\mu$m results in a $\Delta D_{32}$ which is 5% of the $D_{32}$ to be measured. These uncertainties in the $D_{32}$ measurement will cause uncertainties in the $\sigma$ measurements and calculations show that the uncertainties in $\sigma$ measurements are less than 2%. Figure 3.10 shows the results of the uncertainty evaluations at a distribution width of 1.4 and compares the
reconstructed distribution functions with the “true” distribution, which is a log-normal distribution with $D_{32}=3.5 \, \mu m$ and $\sigma=1.4$. These two reconstructed distribution functions represent the bounds of the uncertainties for the SDF measurements by the WMLE scheme described in Figure 3.9 for $D_{32}=3.5 \, \mu m$ and $\sigma=1.4$. Similar results were obtained for other distribution widths. Furthermore, ambiguity in $D_{32}$ measurements ($\Delta D_{32}/D_{32}$) will not exceed that in the case of $D_{32}=3.5 \, \mu m$ (and similarly $D_{32}=1.5 \, \mu m$), where $D_{32}$ could be determined by different wavelength combinations with $\Delta D_{32}$ having opposite signs.

The current availability of laser sources in a wide spectral range from deep UV to the IR supports the feasibility of the WMLE concept depicted in Figure 3.9. For example, a large group of gas lasers operate across the wavelength range from about 200 nm to 12 $\mu m$. The most commonly used devices in this group include lasers based on helium-neon (He-Ne), argon (Ar), and carbon dioxide (CO$_2$). The He-Ne laser can generate wavelengths including 0.5435, 0.6328, 1.523, and 3.391 $\mu m$. The Ar-ion laser can generate wavelengths shorter than the He-Ne laser, such as 257, 488, and 514 nm, and the quadrupled-YAG laser yields light at 266 nm. The CO$_2$ laser lases at longer wavelengths ranging from 9.2 to 11.4 $\mu m$ and from 4.6 to 5.8 $\mu m$ when frequency-doubled. Therefore these primary types of gas lasers can provide or approximate all the wavelengths shown in Figure 3.9. Another group of lasers, semiconductor lasers, provide wavelengths ranging from about 400 nm to 30 $\mu m$ and typically with very low noise and good power levels; especially in the spectral region from 0.6 to 2.0 $\mu m$, these semiconductor lasers can generate virtually any wavelength. Hence the 0.5 and 1.5 $\mu m$ wavelengths used in Figure 3.9 may be readily provided by semiconductor lasers. The semiconductor lasers are also attractive owing to their tunability (i.e., to avoid interference from vapor absorption) and relatively low cost. Besides the gas and semiconductor lasers, other common laser devices commercially available include solid-state and dye lasers whose output wavelengths range typically from about 200 nm to 3.9 mm and about 300 to 750 nm respectively. These laser devices provide a great many
wavelengths in a wide spectral range and will significantly enhance the development of the WMLE technique.

3.5 Extension to Other Aerosol Systems

Discussions in the preceding sections are based on water aerosols at a temperature of 22 °C. This section considers the development of WMLE schemes for the measurement of SDFs for other aerosol systems, including aerosols at different temperatures or aerosols of other liquids such as hydrocarbon fuels. This consideration is essentially the consideration of the impact of refractive indices \( m = n - k \cdot i \) on the wavelength selection process for the WMLE scheme and requires studies of the dependence of extinction coefficients on the refractive indices. Dependence of the \( Q \) and \( \bar{Q} \) curves on the refractive indices is discussed in Appendix C and the major observations are summarized here.

At wavelengths where the imaginary part of the refractive index of the aerosols is not very large (such as for water aerosols in the wavelengths range from 0.25 to 10 μm), the dependence of the \( Q \) and \( \bar{Q} \) curves on the imaginary part can be neglected. Our calculations show that varying the imaginary part of the refractive indices used in this work by ±25% has negligible influence on the wavelength selection process. However, the \( Q \) and \( \bar{Q} \) curves demonstrate a sensitive dependence on the real part of the refractive indices. A small increase in the real part of the refractive index causes the \( Q \) and \( \bar{Q} \) curves to be obviously compressed toward smaller \( D_{32} \), and vice versa. This effect is illustrated in Figure 3.11 and Figure 3.12 by repeating the calculations performed in Figure 3.5 and Figure 3.8 with the real parts of all the refractive indices used in the calculations in Figure 3.5 and Figure 3.8 increased by 5%. Both Figure 3.11 and Figure 3.12 show an overall shift of all the new calculations (performed at
m=1.05n- k·i) toward smaller D_{32} compared with the calculations performed at m=n-k·i. This effect suggests that when applied to aerosols with the real part of the refractive indices larger than that of water aerosols at 22 °C (e.g., water aerosols at a temperature lower than 22 °C or aerosols of some hydrocarbon fuels), the applicable range of the WMLE scheme described in Figure 3.9 is shifted towards smaller D_{32}. On the contrary, when applied to aerosols with real part of the refractive indices smaller than that of water aerosols at 22 °C, the applicable range of this scheme is shifted towards larger D_{12}, which is shown by the calculations performed in Figure 3.11. Also see Appendix C for more discussion.

The impact of the aerosol temperature on the WMLE scheme merits more consideration. The above discussions suggest that SDF measurements by the extinction method depend on the aerosol temperature. This dependence generally renders it necessary to know the aerosol temperature and the refractive indices (especially the real part) at this temperature for the extinction method to measure SDFs accurately. On the other hand, this dependence has the potential to allow non-intrusive measurements of the mean aerosol temperature by the WMLE scheme, which is a critical diagnostic need in many fields of aerosol-related research. These measurements of course require the SDF of the aerosols to be well-characterized and the relationship between the refractive indices and the aerosol temperature to be well understood. The refractive indices of different liquids exhibit different levels of sensitivity to the liquid temperature. For example, the real part of the refractive index of liquid water varies by 0.8% in a temperature range of 20 to 80 °C at wavelengths in the visible range [15, 16]. A similar level of variation is expected at other wavelengths ranging from 0.2 to 10 μm based on the work in [17, 18], except at those wavelengths where a strong absorption band occurs. Such variation in the refractive index is substantially smaller than the variation of 5% considered in Figs. 11 and 12. Therefore, the WMLE scheme shown in Figure 3.9 should allow measurement of SDFs for water aerosols with reasonable accuracy in this temperature range without detailed knowledge of the aerosol temperature. For liquids whose refractive index varies with temperature more drastically.
(e.g., some hydrocarbon fuels [16, 19]), application of the WMLE scheme may require more specific information about the aerosol temperature to ensure the accuracy of the SDF measurements. On the other hand, in such cases the WMLE scheme may have the potential to provide sensitive aerosol temperature measurements. See Appendix C for more detailed calculations and discussions.

### 3.6 Extension to Other Distribution Functions

This section extends the application of the above method for SDF measurements from the log-normal distribution function to other distribution functions. Application of this SDF measurement method to other two-parameter distribution functions (such as the Rosin-Rammler and the normal distribution function) is very similar to that for the log-normal distribution function. Combinations of certain wavelengths are found to demonstrate low sensitivity to the detailed shape of the distribution functions to be measured and high sensitivity to $D_{32}$ for certain ranges of $D_{32}$, and therefore can be used to determine $D_{32}$ in the corresponding range without knowing the exact shape of the distributions. Uncertainties in the determination of $D_{32}$ can be estimated by the methodology described in Section 3 and used to specify the applicable ranges of the wavelength combinations. After $D_{32}$ is measured, the shape of the distribution can be determined by combinations of selected wavelengths at which the ratio of mean extinction coefficients are sensitive to the shape of the distribution, similar to the method discussed in Section 3.4. But note that, as pointed out earlier in this chapter, a priori information about the form of the SDFs to be measured must be known.

Here we focus our discussion on the extension of this SDF measurement method to the upper limit distribution function (ULDF) as introduced in [20], which is a three-parameter distribution function defined as follows
\[
f(D) = B \frac{\exp\left[-(\delta \ln \frac{aD}{D_\infty - D})^2\right]}{D^3(D_\infty - D)}
\]

(3.11)

where:

\(a\) and \(\delta\): the shape parameters of the distribution

\(D_\infty\): the largest diameter in the distribution

\(B\): a normalization constant such that the distribution function is normalized, i.e.

\[\int_0^{D_\infty} f(D) dD = 1.\]

The three parameters in the ULDF (\(a\), \(\delta\), and \(D_\infty\)) can be adjusted independently to vary the mean size, distribution width, and the skewness of the distribution. Figure 3.13 shows four ULDFs at various \(a\) and \(\delta\)’s (the values of the \(a\) and \(\delta\)’s are listed in the caption) with the same \(D_\infty\). Since the shape parameters (\(a\) and \(\delta\)) give no apparent interpretation for the geometrical form of the distributions, Deiss [21] suggested the use of two nondimensional factors, a width factor (\(W_D\)) and a skewness factor (\(S\)) as defined in Equation (3.12) and (3.13), to describe the ULDFs

\[W_D = \omega / \bar{D}\]

(3.12)

\[S = \bar{D} / D_\infty\]

(3.13)

where:

\(\bar{D}\) = the most probable diameter in the distribution as shown in Figure 3.13

\(\omega\) = the width of the distribution at the two diameters (\(D^+\) and \(D^-\) in Figure 3.13) where the value of \(f(D)\) is half that of \(f(\bar{D})\)

According to the definition in Equation (3.13), a larger \(S\) corresponds to a distribution with modal diameter closer to \(D_\infty\) and vice versa. We modify the definition of the width factor defined in Equation (3.12) to the following one

\[W = \omega / D_\infty\]

(3.14)
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Obviously W is simply the product of \( W_D \) and S, however a larger W defined in Equation (3.14) always corresponds to a distribution with wider halfwidth and vice versa, which is not true for the \( W_D \) defined in Equation (3.12). Either the shape parameters (a and \( \delta \)) or the nondimensional factors (W and S) can fully specify the shape of a ULDF and this work uses the nondimensional factors. The nondimensional width and skewness factor are related to the shape parameters through a set of transcendental equations [22], and they are calculated for the distributions presented in Figure 3.13. The geometrical forms of the distributions displayed in Figure 3.13 suggest that ULDFs with S and W in the range of 0.2 to 0.8 and 0.15 to 0.5, respectively, represent a relatively wide class of distributions in terms of distribution width and skewness. Therefore, here we limit our discussion to ULDFs with S and W in the ranges of 0.2 to 0.8 and 0.15 to 0.5, respectively.

Finally, the Sauter mean diameter (\( D_{32} \)) of a ULDF is related to the parameters of the distribution by the following equation [20]

\[
D_{32} = \frac{D_\infty}{1 + a \exp(1/4\delta^2)}
\]  

(3.15)

Figure 3.14 shows the calculations of mean extinction coefficients (\( \bar{Q} \)) at three wavelengths for water aerosols following the upper limit distributions specified in Figure 3.13. These extinction coefficient curves exhibit similar profiles as those shown in Figure 3.2, indicating the discussions and analysis for log-normal distributions will apply for ULDFs. Similar to the observations made in Figure 3.2, \( \bar{Q} \) at each wavelength shows low sensitivity to the shape of the distribution functions on certain \( D_{32} \) ranges, and the same analysis performed in Section 3.2 applies here to explain this low sensitivity. For example, the variations in \( \bar{Q} \) at 10 \( \mu \)m remain small over the entire \( D_{32} \) range shown in Figure 3.14, even if the distributions have quite different shapes, and the variations in \( \bar{Q} \) at 0.5 \( \mu \)m starts to be damped out after a \( D_{32} \) of 4 \( \mu \)m. Therefore, the ratio (R) between \( \bar{Q} \) at 10 and at 0.5 \( \mu \)m (R) will be insensitive to the shape of the
distributions and can be used to measure $D_{32}$ ranging from 4 to 12 µm without detailed knowledge of the shape parameters (or equivalently, the nondimensional factors) of the distribution. Ambiguity will occur in the determination of $D_{32}$ because of the lack of knowledge about the shape parameters, and this ambiguity is still quantified by $\Delta D_{32}/D_{32}$ defined in Equation (3.8). However, here the definition of $D_{32}^1$ and $D_{32}^2$ need to be modified to be the largest and smallest possible $D_{32}$ determined by $R$ curves between a pair of wavelengths in the $W$ and $S$ ranges under consideration, and $\Delta D_{32}/D_{32}$ is always positive under these modified definitions.

Figure 3.15 shows the calculation of $\Delta D_{32}/D_{32}$ when $R$ between $\bar{Q}$ at 10 and 0.5 µm is used to determine $D_{32}$ for water aerosols following upper limit distributions with $W$ and $S$ in the range of 0.15 to 0.5 and 0.2 to 0.8 respectively. As expected from the above analysis of $\bar{Q}$, the combination of these two wavelengths enables measurements of $D_{32}$ within 5% for $4.2 < D_{32} < 12$ µm. Other wavelengths can be selected to measure $D_{32}$ in other ranges. For example, Figure 3.15 shows that a wavelength combination of 4 and 0.5 mm enables determination of $D_{32}$ within 5% for $2.6 < D_{32} < 4.2$ µm, and a combination of 1.75 and 0.25 µm for $1.3 < D_{32} < 2.6$ µm. Similar to the discussions of the measurements of $D_{32}$ for log-normal distributions, more wavelengths can be incorporated to enable the measurement of $D_{32}$ with smaller ambiguity and to expand the measurements to wider ranges of $D_{32}$, $W$, and $S$. Moreover, the influence of refractive index on the $D_{32}$ measurements also has the same trend as that analyzed in the case of log-normal distributions.

However, the determination of the shape parameters of the ULDFs after $D_{32}$ has been measured requires a different method than that developed for the two-parameter distributions. The reason is that no such wavelength can be found at which $\bar{Q}$ is sensitive to one of the shape parameters but insensitive to the other, which is illustrated by calculations of $\bar{Q}$ in Figure 3.14. Consequently, the influence of the shape parameters on $\bar{Q}$ cannot be decoupled and determined.
separately. An optimization method [14] or a matrix inversion method [4] can be applied here to solve for the shape parameters. But the methodology developed in Section 4 for the evaluation of sensitivity of $\Omega$ to the shape parameters still applies here and would provide useful guidance to determine the application ranges of different wavelengths to achieve optimum measurement sensitivity and accuracy.

### 3.7 Summary

Strategies based on WMLE for SDF measurements have been introduced and developed for a variety of distribution functions. Incorporation of wavelengths from a wide spectral range in the WMLE scheme was demonstrated to enable a stable inversion of the extinction measurements to SDFs with enhanced accuracy. Sensitivity analysis reveals the proper ranges over which the extinction measurements at the wavelengths selected in the WMLE scheme should be applied for optimized sensitivity and accuracy of the SDF measurements. These advantages are illustrated by an example WMLE scheme employing wavelengths in the spectral range from 0.25 to 10 µm to measure SDFs of water aerosols. Application of this approach to other aerosol systems is also considered. The WMLE scheme was found to provide stable determination of a variety of SDFs with Sauter mean diameters ranging from sub-micron to about 10 µm.

In summary, this work developed a systematic methodology for evaluating and selecting wavelengths from a wide spectral range (0.25 to 10 µm) for droplet sizing using laser extinction, and analyzed the sensitivity and applicable range of different wavelengths for different spray systems and distribution functions. Based on these analyses, a relatively simple yet stable algorithm compared to previous work is developed to invert the extinction data into SDFs. The analysis performed here should provide theoretical support for the practical implementation of the WMLE concept, and motivate further research work on the development of this concept.
Moreover, by incorporation of additional wavelengths to monitor the spectral absorption by the vapor in association with the aerosols, the WMLE scheme can be extended to enable simultaneous measurements of multiple quantities both of the aerosols and of the vapor, including the SDF and loading of the aerosols, and the concentration and temperature of the vapor. Such extensions will be discussed in the next chapter.

References:


Figure 3.1. Log-normal distribution functions with $D_{32}=4 \, \mu m$ and different distribution widths ($\sigma$).

Figure 3.2. $\bar{Q}$ at different wavelengths for water aerosols at a temperature of 22 °C following log-normal distribution with various distribution widths ($\sigma$'s). Refractive indices are taken to be $m= 1.335$ at $\lambda=0.5 \, \mu m$, $m= 1.351-0.0046i$ at $\lambda=4 \, \mu m$, and $m=1.218-0.0508i$ at $\lambda=10 \, \mu m$. 
Figure 3.3. Ratio between $\bar{Q}$ at 10 and 0.5 $\mu$m (i.e. $R = \frac{\bar{Q}(10 \mu m)}{\bar{Q}(0.5 \mu m)}$) shown in Figure 3.2 at three distribution widths ($\sigma$'s). The inset illustrates the ambiguity ($\Delta D_{32}$) in $D_{32}$ determination due to the lack of detailed knowledge about the distribution width.

Figure 3.4. Ambiguity in the determination of $D_{32}$ ($\Delta D_{32}/D_{32}$) by the R curves shown in Figure 3.3 for different ranges of distribution widths ($\sigma$'s) to specify the applicable range for the wavelength combination of 0.5 and 10 $\mu$m for $D_{32}$ measurements.
Figure 3.5. Wavelengths selected in the WMLE scheme for the measurement of $D_{32}$ of water aerosols following log-normal distributions with distribution widths ($\sigma$'s) ranging from 1.2 to 1.6.

Figure 3.6. Ratio between $Q$ at 4 and 0.5 $\mu$m shown in Figure 3.2 at three distribution widths ($\sigma$'s) to illustrate that $R$ shows different sensitivity to the distribution width over different ranges of $D_{32}$. 
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**Figure 3.7.** Variations of $R$ between $\bar{Q}$ at 0.5 and 4 $\mu$m shown in Figure 3.2 at two selected $D_{32}$’s for distribution width in the range of 1.2 to 1.6 and the sensitivity analysis of this ratio for distribution width measurements.

**Figure 3.8.** Calculations of $S_{\sigma}^{\text{min}}$ for wavelengths selected in the WMLE scheme to specify the applicable $D_{32}$ ranges of different wavelength combinations for $\sigma$ measurement with optimized sensitivity.
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Figure 3.9. Summary of the wavelengths selected in the WMLE scheme to measure SDFs of water aerosols following log-normal distributions, the applicable D_{32} range for each of the wavelength combinations, and the criteria considered in the wavelength selection process.

Figure 3.10. Uncertainty analysis for SDF measurements by the WMLE scheme described in Figure 3.9, i.e., $\lambda=4$ and 0.5 $\mu$m are combined to determined $D_{32}$ and $\lambda=10$ and 4 $\mu$m are combined to determined $\sigma$. 

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Figure 3.11. Evaluation of the influence of refractive indices on the applicable ranges of the WMLE scheme for $D_{32}$ measurements. Here, $n$ and $k$ represent the real and imaginary part of the refractive index at the corresponding wavelengths as specified in Figure 3.2.

Figure 3.12. Evaluation of the influence of refractive indices on the applicable ranges of the WMLE scheme for distribution width measurements.
Figure 3.13. Four example ULDFs with different skewness and widths. The shape parameters are $a=0.25$ and $\delta=0.88$ for $S=0.8$ and $W=0.5$, $a=0.25$ and $\delta=1.89$ for $S=0.8$ and $W=0.15$, $a=0.57$ and $\delta=0.88$ for $S=0.2$ and $W=0.5$, $a=2.63$ and $\delta=1.89$ for $S=0.2$ and $W=0.15$.

Figure 3.14. $Q$ at different wavelengths for water aerosols at a temperature of 22 °C following the ULDFs specified in Figure 3.13.
Figure 3.15. Wavelengths selected for the measurement of $D_{32}$ of water aerosols following ULDFs with W and S in the range of 0.15 to 0.5 and 0.2 to 0.8 respectively.
Chapter 4

Simultaneous Vapor and Droplet Measurement Using WMLE

4.1 Introduction

Multiphase flows involving liquid droplets in association with a gas flow occur in many industrial and scientific applications. In such applications, it would be desirable to monitor both the vapor and the droplets simultaneously.

Numerous laser-based techniques [1-6] have been developed for the separate characterization of either vapor or droplets as discussion in Chapter 1. However, the fact that scattering from droplets greatly exceeds that from the vapor in most applications discourages the direct combination of existing vapor and droplet sensing techniques for the simultaneous monitoring of both vapor and droplet properties. Alternative approaches for simultaneous vapor and droplet measurement are therefore sought.

Past work on this topic includes Laser Induced Exciplex Fluorescence (LIEF) developed by Melton [7] and a two-wavelength laser extinction strategy proposed by Chraplyvy and Tishkoff [8, 9]. However, the LIEF measurements are difficult to quantify. The techniques based on laser extinction are quantitative and offer several key advantages for applications in propulsion systems as discussed in proceeding chapters.
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In this chapter, we extend the vapor and droplet sensing techniques introduced in Chapter 2 and 3 to a new diagnostic concept based on wavelength-multiplexed laser extinction (WMLE) to enable simultaneous characterization of droplet properties (droplet size, loading, etc.) and gas properties (gas concentration, temperature, etc.). The first extension considered incorporates a vapor sensing technique modified from spectrally-resolved absorption described in Chapter 2, namely differential absorption, to significantly improve the detection limit of vapor sensing. The second extension considered includes laser sources at an arbitrary number of wavelengths in a wide spectral range to expand sensing capability of the diagnostic to more quantities besides vapor concentration and droplet volume fraction. Systematic methodologies are developed to guide the selection of wavelengths to achieve optimized performance for both vapor and droplets measurement. An example WMLE scheme utilizing wavelengths ranging from 0.5 to 10 µm is illustrated for droplet size and vapor concentration measurements through an example of water spray, and are found to enable unique and sensitive Sauter mean diameter measurement in the range of ~1 to 10 µm along with accurate vapor detection.

4.2 Theory

When a collimated light beam at wavelength $\lambda$ is incident upon a mixture of vapor and droplets with uniform properties along the probing pathlength, the transmitted beam intensity ($I_t$) is related to the incident beam intensity ($I_0$) by

$$T(\lambda) = \frac{I_t}{I_0} = \exp[-\tau_{vapor}(\lambda)] \cdot \exp[-\tau_{drops}(\lambda)]$$

$$= \exp[-\alpha(\lambda) \cdot X \cdot P \cdot L] \cdot \exp[-\tau_{drops}(\lambda)]$$

(4.1)

where:

$T(\lambda)$ = the optical transmittance at wavelength $\lambda$
\[ \tau_{\text{vapor}}(\lambda) = \text{the vapor absorbance} \]
\[ \tau_{\text{drops}}(\lambda) = \text{the droplet extinction} \]
\[ \alpha(\lambda) = \text{the absorption coefficient of vapor (cm}^{-1}\cdot\text{atm}^{-1}) \]
\[ X = \text{the vapor mole fraction} \]
\[ P = \text{the total (mixture) pressure along the pathlength (atm)} \]
\[ L = \text{the pathlength (cm)} \]

Absorption by vapor has been discussed in Chapter 2. At wavelengths where vapor absorption does not occur (i.e., \( \alpha(\lambda) = 0 \)), attenuation of the incident beam is only due to extinction from the droplet, which has been discussed in Chapter 3. For the convenience of discussion in this chapter, light scattering theory is briefly reviewed here and several new definitions and notations will be introduced.

From light scattering theory [10], extinction by a collection of mono-dispersed spherical droplets is related to the number density and size of the droplets in the following equation

\[
\tau_{\text{drops}} = C_n \cdot \frac{\pi D^2}{4} \cdot Q(\frac{\pi D}{\lambda}, m) \cdot L 
\]

(4.2)

where:
\[ C_n = \text{the number density of the droplets (cm}^{-3}) \]
\[ D = \text{diameter of the droplets (cm)} \]
\[ Q(\frac{\pi D}{\lambda}, m) = \text{the extinction coefficient of a droplet with diameter D at wavelength } \lambda \]
\[ m = \text{the complex refractive index of the droplets at wavelength } \lambda \]
\[ L = \text{pathlength} \]

\( Q(\pi D/\lambda, m) \) is a complex function of the incident wavelength and droplet diameter and calculation of \( Q(\pi D/\lambda, m) \) requires numerical methods except in some special cases. This work uses a Mie scattering algorithm described in [11] to compute the droplet extinction coefficients. In the case of a collection of poly-dispersed droplets, Equation (4.2) needs to be modified to
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\[
\tau_{\text{drops}} = \frac{\pi}{4} C_n L \int_0^\infty Q(\pi D / \lambda, m) f(D) D^2 dD
\]  

(4.3)

where \( f(D) \) is the droplet size distribution function defined such that \( \int_0^\infty f(D) dD = 1 \), and \( f(D)dD \) represents the probability that a droplet has diameter between \( D \) and \( D+dD \). Due to the lack of a fundamental mechanism or model to build droplet size distribution functions theoretically, various distribution functions have been used based on probability analysis or empirical observations. A log-normal function, defined as following, is one of the most commonly used distribution functions

\[
f(D) = \frac{1}{\sqrt{2\pi D \ln \sigma}} \exp\left[-\frac{1}{2(\ln \sigma)^2} (\ln D - \ln \overline{D})^2\right]
\]  

(4.4)

In this equation, \( \sigma \) represents the standard deviation of the distribution (distribution width), \( \overline{D} \) the count median diameter, which means 50% of the droplets in the distribution have diameter smaller than \( D_{\text{CM}} \) and 50% larger than \( \overline{D} \).

In many calculations and applications, it is convenient to restrict the work to averaged parameters instead of using the complete size distribution function. Based on \( f(D) \), many statistical parameters about the droplet distribution can be defined. Two useful parameters, the mean scattering coefficient \( \overline{Q} \) and the Sauter Mean Diameter \( (D_{32}) \), are defined as following

\[
\overline{Q} = \frac{\int_0^\infty Q(\pi D / \lambda, m) f(D) D^2 dD}{\int_0^\infty f(D) D^2 dD}
\]  

(4.5)

\[
D_{32} = \frac{\int_0^\infty f(D) D^3 dD}{\int_0^\infty f(D) D^2 dD}
\]  

(4.6)

Obviously, for a given droplet distribution, \( \overline{Q} \) depends on the incident wavelength and some characteristic diameter of the droplets. \( D_{32} \) is used as such a characteristic diameter in this
paper and this dependence is denoted as $\bar{Q}(\pi D / \lambda, D_{32})$. Figure 4.1 provides some example calculations to show the variations of $Q$ (for mono-dispersed droplets) and $\bar{Q}$ (for poly-dispersed droplets) with droplet size at three wavelengths for water droplets. In the case of poly-dispersed droplets, size distribution functions are taken to be the log-normal function defined in Equation (4.4) with different distribution widths, as illustrated in the insert of Figure 4.1. Refractive indices used in the calculations shown in Figure 4.1 are from measurements made in [12, 13]. An important observation is that the positions of principal maxima of the extinction coefficient curve display only a weak dependence on the width ($\sigma$) of the log-normal distribution functions. Furthermore, a weak dependence on the distribution functions is found both in our work and in studies elsewhere [14], especially when the distribution width (characterized by the standard deviation of the distribution function) is large. Also note the insensitivity of extinction coefficients to distribution width at a wavelength of 10 $\mu$m.

Finally, by introducing a new parameter, the droplet volume fraction ($C_V$, defined as ratio between the volume occupied by the droplets and the total volume), Equation (4.2) and (4.3) can be transformed into the following forms

$$
\tau_{\text{drops}} = \frac{3C_v}{2D} \bar{Q}(\pi D / \lambda, m)L \quad \text{for mono-dispersed droplets} \quad (4.7)
$$

and

$$
\tau_{\text{drops}} = \frac{3C_v}{2D_{32}} \bar{Q}(\pi D_{32} / \lambda, m)L \quad \text{for poly-dispersed droplets} \quad (4.8)
$$

where $C_V$ is the volume fraction occupied by liquid droplets along the pathlength, and can be calculated by the following equations

$$
C_V = C_n \cdot \frac{\pi}{6} D^3 \quad \text{for mono-dispersed droplets} \quad (4.9)
$$

and

$$
C_V = C_n \int_0^\infty \frac{\pi}{6} D^3 f(D) dD \quad \text{for poly-dispersed droplets} \quad (4.10)
$$

$C_n$ in Equation (4.9) and (4.10) is the number density of the droplets along the pathlength.
4.3 Droplet Measurement

As described in Chapter 3, droplets (size distribution, mean diameter, and volume) can be characterized by extinction measurements at multiplexed wavelengths. More specifically, the ratios of extinction measurements at different wavelengths as shown in Equation (4.11) are used to determine the mean diameter and size distribution of the droplets. Then the absolute extinction measurements are used to determine the volume fraction of the droplets.

\[
\frac{\tau_{\text{drops}}(\lambda_1)}{\tau_{\text{drops}}(\lambda_2)} = \frac{Q(\pi D_{32} / \lambda_1, m)}{Q(\pi D_{32} / \lambda_2, m)} = R
\]  

(4.11)

Based on the study of the extinction ratios between different wavelengths, a WMLE scheme was designed to monitor water droplets in Chapter 3. Here, the criteria considered in the design are reviewed to provide a systematic method for the selection of wavelengths in WMLE scheme for droplet measurement.

**Criterion A.** As discussed in Chapter 3, combination of different wavelengths results in different level of accuracy and sensitivity in the measurement of mean size and distribution width of the droplets. Therefore, this criterion requires that the selected wavelengths provide sensitive determination of droplet size and distribution shape over the range of interest. In this work, sensitivity is quantified by \( S \) as defined below.

\[
S = \left| \frac{dR / R}{dD_{32} / D_{32}} \right| 
\]  

(4.12)

This definition has been introduced and discussed in Chapter 3. A large \( S \) implies that a small proportional change in \( D_{32} \) results in large proportional change in \( R \), therefore sensitive determination of \( D_{32} \) is enabled.

**Criterion B.** For each combination of two wavelengths, the ratio of extinction coefficients (\( R \)) is monotonic only over a certain range of droplet diameter (\( D_{32} \)) as shown in
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Figure 4.2. For a D_{32} outside this range, this combination of two wavelengths becomes insufficient to provide a unique determination of D_{32}. Criterion B requires that the selected wavelengths provide monotonic R over the range of droplet size to be measured to enable unique determination of droplet size.

**Criterion C.** For each combination of two wavelengths, the ratio of extinction coefficients (R) is very small over a certain range of D_{32}. According to Equation (4.11), very small R implies either very strong or very weak extinction at one of the wavelengths. However both very strong extinction and very weak extinction impair signal to noise ratio (SNR) of the measurement. A transmittance (I/I_0) between 0.4 and 0.9 is recommended for optimum extinction measurement [18]. Based on this recommendation and Equation (4.1), the optimum R for droplet sizing should lie between 0.1 and 10. Therefore, Criterion C requires that the ratio of extinction coefficients at the selected wavelengths be between 0.1 to 10 to ensure optimum SNR over the range of droplet size to be measured.

Design of WMLE scheme considering all these criteria was illustrated in Chapter 3, with an example scheme shown in Figure 3.9.

### 4.4 Vapor Concentration Measurement

#### 4.4.1 Concept

Theoretically, after the contribution from droplet extinction is known from the droplet measurement, the vapor concentration could be obtained from the measurement of extinction at a wavelength where vapor absorbs [8, 9, 19]. However, the accuracy of this method is jeopardized when the contribution from droplet extinction is larger than that from vapor absorption (i.e., $\tau_{\text{drops}} > \tau_{\text{vapor}}$) even when $\tau_{\text{vapor}}$ itself is large enough to allow accurate vapor measurement, therefore
limiting the applicable range of this method. An alternative strategy has been sought to extend the vapor detection range.

Our approach for vapor sensing is based on a differential absorption concept. This concept takes advantage of the observation that vapor absorption varies much more rapidly with wavelength than the droplet extinction, based on the fact that vapor spectral features are spectrally narrower than corresponding liquid spectra.

Figure 4.3 illustrates this concept schematically, with $\lambda_3$ as the vapor detection wavelength. When extinction from droplets exceeds that from vapor absorption as shown in part a) of Figure 4.4 (i.e. $\tau_{\text{drop}}(\lambda_3) > \tau_{\text{vapor}}(\lambda_3)$), vapor measurements at this wavelength suffer by interference from strong droplet extinction. However, as shown in part b) of Figure 4.3, the differential vapor absorption between $\lambda_3$ and another wavelength $\lambda_4$ (defined as $\Delta\tau_{\text{vapor}} = |\tau_{\text{vapor}}(\lambda_3) - \tau_{\text{vapor}}(\lambda_4)|$), can be comparable or larger than the differential droplet extinction (defined as $\Delta\tau_{\text{drops}} = |\tau_{\text{drop}}(\lambda_3) - \tau_{\text{drop}}(\lambda_4)|$), therefore extending accurate vapor detection into a wider dynamic range.

### 4.4.2 Selection of Wavelengths

Obviously the selection of wavelengths ($\lambda_3$ and $\lambda_4$) to optimize vapor detection depends on the spectra of the specific vapor and liquid under consideration. This section again uses water as an example to illustrate a methodology for wavelength selection, but the approach is also applicable to other vapor and liquid systems.

We first examine the dependence of differential droplet extinction ($\Delta\tau_{\text{drops}}$) on selected wavelengths ($\lambda_3$ and $\lambda_4$) with the goal of minimizing differential droplet extinction. Substitution of Equation (4.7) (from mono-dispersed droplets) into the definition of $\Delta\tau_{\text{drops}}$ yields

$$\Delta\tau_{\text{drops}} = \frac{3C_y |Q(\lambda_3, D) - Q(\lambda_4, D)| L}{2D} = \frac{3C_y L}{2D} \Delta Q \quad (4.13)$$
Therefore the dependence of $\Delta \tau_{\text{drops}}$ on selected wavelengths is equivalent to the dependence of $\Delta Q$ (i.e. the differential extinction coefficient) on selected wavelengths.

Figure 4.4 shows the droplet extinction coefficient and vapor absorption over wavelength in a spectral range from 0.5 to 9 $\mu$m for water vapor and droplets with two diameters, using the liquid spectra data of water measured in [12, 13], and the vapor spectra is simulated using HITRAN 2004. Note the relatively slow variation of extinction coefficient compared to that of vapor with wavelength, as mentioned before. Examination of $\Delta Q$ at different combinations of $\lambda_3$ and $\lambda_4$ within this spectral range shows a general profile of $\Delta Q$ similar to that shown in Figure 4.5, where $\lambda_3$ is chosen to be 5 $\mu$m.

Figure 4.5 reveals that multiple combinations of wavelengths can be applied to achieve small $\Delta Q$ for a given droplet diameter. For example, for a droplet diameter of 6 $\mu$m and a $\lambda_3$ of 5 $\mu$m, either a $\lambda_4$ near 5 or 3 $\mu$m results in small $\Delta Q$. These wavelength combinations can be divided into two categories. The first category is that a combination of two wavelengths close to each other (i.e. $\lambda_3 = 5 \mu$m and $\lambda_4$ close to $5 \mu$m in the above example), and the second is that a combination of two wavelengths well separated from each other (i.e. $\lambda_3 = 5 \mu$m and $\lambda_4$ close to $3 \mu$m in the above example). However the second category of wavelength combinations is only applicable to cases with small droplet size changes. As shown in Figure 4.6, when droplet diameter changes from 6 to 3 $\mu$m, $\Delta Q$ increases by a large amount if $\lambda_4$ is well separated from $\lambda_3$, while small $\Delta Q$ is maintained if $\lambda_4$ is close to $\lambda_3$. Mathematical considerations of the dependence of $Q$ on droplet size and wavelength validate the generalization of above observations. Therefore in general, wavelengths for vapor measurement by differential absorption must be close to each other for applications with varying droplet size.

Next we consider the dependence of differential vapor absorption ($\Delta \tau_{\text{vapor}}$) on selected wavelengths ($\lambda_3$ and $\lambda_4$) with the goal of maximizing differential vapor absorption. Apparently
maximum differential absorption is achieved by selecting one wavelength (e.g. $\lambda_3$) where the strongest vapor absorption occurs and another wavelength (e.g. $\lambda_4$) where the weakest vapor absorption occurs over the entire spectral range of interest. For the example of water as shown in Figure 4.4, one wavelength ($\lambda_3$) can be at a vapor absorption peak near 2.7 or 5.9 µm. Part a) of Figure 4.6 shows the selection of $\lambda_3$ at 2.6705 µm, where maximum vapor absorbance occurs in the 2.5 to 3.0 µm spectral range. For the case with varying droplet size, which is of major interest to this work, the second wavelength ($\lambda_4$) must be close to $\lambda_3$. Part b) of Figure 4.6 shows water vapor spectra in the vicinity of 2.6705 µm. In principle, a $\lambda_4$ chosen to be as close to $\lambda_3$ as possible provided that vapor does not absorb at this wavelength can minimize $\Delta \tau_{\text{drops}}$ while maintaining maximum $\Delta \tau_{\text{vapor}}$. A $\lambda_4$ of 2.6720 µm as shown in part b) of Figure 4.6 is an example of such a choice. But in practice, other considerations such as avoiding interference from pressure broadening of the vapor spectra may encourage a different selection of $\lambda_4$, for example, 2.6818 µm as shown.

Finally we evaluate the applicable range of the above differential scheme for water vapor detection. Consider a collection of water droplets (with a monodispersed size distribution) in an inert bath gas undergoing uniform evaporation at constant pressure and temperature, as depicted in Figure 4.7. The system starts at time=0 with all droplets at diameter $D_0$, droplet volume fraction $C_{V0}$, and no vapor. After a time of t, the diameter of the droplets decreases to D due to evaporation and consequently the droplet volume fraction decreases to $C_V$ and the mole fraction of water vapor in the system increases to $X$. Finally at time=T, all the droplets are evaporated. The differential scheme designed above with $\lambda_3$ at 2.6705 µm and $\lambda_4$ at 2.6818 µm is applied to monitor vapor concentration as shown. Comparison between $\Delta \tau_{\text{drops}}$ and $\Delta \tau_{\text{vapor}}$ during the evaporation process provides an evaluation of the applicable range of this vapor sensing scheme in terms of droplet size (D), droplet loading ($C_V$), and mole fraction of vapor ($X$). This work uses
the range where $\Delta \tau_{\text{vapor}}$ exceeds $\Delta \tau_{\text{drops}}$ to quantify the applicable range of the differential absorption scheme.

This comparison is complicated because it depends on both initial droplet diameter ($D_0$) and droplet volume fraction ($C_{V_0}$). Comparison at some representative initial diameter is therefore used to provide an insight into the applicable range of the differential absorption scheme. One such representative diameter can be obtained by seeking a diameter to maximize differential droplet extinction ($\Delta \tau_{\text{drops}}$) in Equation (4.13) for a given combination of differential absorption wavelengths. This initial diameter, denoted as $D_0^{\text{max}}$, results in maximum $\Delta \tau_{\text{drops}}$ at time=0 in the process depicted in Figure 4.7. Furthermore, both mathematical considerations and numerical computations reveal that other initial diameters will yield similar or smaller $\Delta \tau_{\text{drops}}$ during the entire evaporation process compared to that yielded at $D_0^{\text{max}}$. Therefore, comparison between $\Delta \tau_{\text{drops}}$ and $\Delta \tau_{\text{vapor}}$ at $D_0^{\text{max}}$ provides a lower limit of the applicable range of the differential absorption, i.e. the applicable range for other initial droplet diameters will be similar or wider than that evaluated at $D_0^{\text{max}}$. For the differential scheme shown in Figure 4.6 ($\lambda_3$ at 2.6705 and $\lambda_4$ at 2.6818 $\mu$m), $D_0^{\text{max}}$ is found to be 5.1 $\mu$m. Therefore, the range where $\Delta \tau_{\text{vapor}}$ exceeds $\Delta \tau_{\text{drops}}$ evaluated at $D_0^{\text{max}}=5.1$ $\mu$m represents a lower limit of the applicable range of this differential absorption scheme, and this scheme can be applied for vapor detection in a wider range at other $D_0$’s.

Vapor absorption and droplet extinction at a single wavelength ($\lambda_3=2.6705$ $\mu$m) are first calculated in Figure 4.8 for a few initial droplet volume fractions at $D_0=5.1$ $\mu$m. Note a $C_{V_0}$ of 19.36 ppm (parts per million) corresponds to the case that the saturated water vapor pressure at a temperature of 22 °C is reached when all the droplets in the system shown in Figure 4.7 are evaporated. Therefore at a $C_{V_0}$ higher than 19.36 ppm (for example 40 ppm), the saturated water vapor pressure is reached when the droplets are only partially evaporated (about 50% evaporated
in the example of \( C_{V0} = 40 \text{ ppm} \), and vapor absorption becomes a constant afterwards. Figure 4.8 suggests that accurate vapor measurement by single wavelength is jeopardized by large droplet extinction interference for some range of droplet volume fraction while vapor absorption itself is strong enough to allow accurate vapor measurement. For example, for the case when \( C_{V0} = 40 \text{ ppm} \), after about 10% of the droplets are evaporated, the vapor absorbance reaches about 10% and accurate vapor detection can be readily performed at this absorbance level. However, droplet extinction imposes significant interference on vapor detection because \( \tau_{\text{drops}} > \tau_{\text{vapor}} \) by almost a factor of three at this stage of evaporation as shown in Figure 4.8. Once at least 22% of the droplet volume evaporates (i.e. \( C_V/C_{V0} < 78\% \)), \( \tau_{\text{vapor}} \) starts to exceed \( \tau_{\text{drops}} \), and this single wavelength detection scheme becomes applicable afterwards by the definition in this work. Therefore, the fact that droplet extinction exceeds vapor absorption in a certain range limits the applicable range of direct absorption for measurement of vapor concentration.

The differential absorption strategy extends the vapor measurement into the range where strong interference from droplet extinction impairs the utility of single wavelength detection. Figure 4.9 shows the comparison between \( \Delta \tau_{\text{drops}} \) and \( \Delta \tau_{\text{vapor}} \) when the differential scheme (\( \lambda_3 \) at 2.6705 and \( \lambda_4 \) at 2.6820 \( \mu \text{m} \)) is applied to the process described in Figure 4.7. Obviously \( \Delta \tau_{\text{vapor}} \) starts to exceed \( \Delta \tau_{\text{drops}} \) much earlier in the process than \( \tau_{\text{vapor}} \) starts to exceed \( \tau_{\text{drops}} \) in Figure 4.8, therefore a wider domain of accurate vapor detection is achieved using differential absorption. In the case shown in Figure 4.9, vapor detection is limited by the magnitude of vapor absorption itself, not by interference from droplet extinction as in the case of Figure 4.8. For example, for the case when \( C_{V0} = 40 \text{ ppm} \), after about 2% of the droplets are evaporated (i.e. \( C_V/C_{V0} < 98\% \)), vapor absorbance starts to exceed droplet extinction and therefore the differential absorption scheme becomes applicable for vapor detection afterward. Although differential vapor absorbance is low (\(~1.5\%) because the vapor concentration is low at this stage of evaporation, stronger differential signal can be achieved by utilizing a longer pathlength. Note that a \( D_0 \) of 3.0 \( \mu \text{m} \) results in
smaller $\Delta \tau_{\text{drops}}$ than that from a $D_0$ of 5.1 µm as expected. Also note that the applicable range of differential absorption is not strongly sensitive to normalized droplet volume fraction (i.e. $C_V/C_{V0}$) from Figure 4.9.

### 4.4.3 Differential Absorption and Wavelength Availability

Discussions in Section 4.4.2 are based on the assumption that light sources are available at any wavelength to allow the utilization of the strongest differential vapor absorption. Although the strongest differential vapor absorption cannot be always accessed in practice due to the lack of light sources at proper wavelengths, it can be shown that differential absorption always extends vapor detection into wider domain compared with the single wavelength technique. Major conclusions about wavelength selection are transferable to other droplet/vapor systems than water and the methodology demonstrated above can be used evaluate the applicable range of specific differential absorption scheme. For example, the wavelengths near 2.67 µm shown in Figure 4.4 can be replaced by readily-available diode laser sources near 1.39 µm, where the strongest absorption of the $\nu_1+\nu_3$ band of water vapor occurs. Though the absorption from water vapor near 1.39 µm is weaker than that near 2.67 µm by about an order of magnitude, a differential absorption scheme based on the absorption features near 1.39 µm still enables $\Delta \tau_{\text{vap}}$ to start to exceed $\Delta \tau_{\text{drops}}$ in an early stage of evaporation in the process defined in Figure 4.7, and therefore allow accurate vapor detection in a wide range of droplet loading and vapor concentration.

### 4.5 Summary

Strategies for droplet and vapor measurement based on laser extinction have been introduced in section 4.3 and 4.4 respectively. Combination of these strategies can be realized by wavelength multiplexing. The Wavelength-Multiplexed Laser Extinction (WMLE) scheme
shown in Figure 4.10. represents an implementation of such combination. Additional laser sources at an arbitrary number of wavelengths can be incorporated into this scheme to enable simultaneous sensing of more parameters. For example, if an additional wavelength ($\lambda_5$ in Figure 4.10) is added to monitor vapor absorption, then vapor temperature can be extracted using the ratio of vapor absorption monitored at $\lambda_4$ and $\lambda_5$ [3], or ratio of integrated absorbance areas if the vapor absorption spectra can be scanned quickly by tunable laser sources [20].

Implementation of the WMLE scheme depicted in Figure 4.10 will be discussed in the next chapter to obtain real-time simultaneous measurement of droplet size, volume fraction, vapor concentration, and vapor temperature in a multiphase flow containing evaporating water droplets. This WMLE scheme consisting of laser sources spanning from visible to mid-IR was devised and tested in an aerosol shock tube facility to study the water droplet evaporation behind shock waves. Among these wavelengths, five wavelengths, 0.63, 1.62, 2.05, 3.39, and 10.0 $\mu$m, were used to measure the droplet size, droplet volume fraction, and distribution function of the evaporating water droplets; and another three wavelengths near the 1.39 $\mu$m spectral region were used to monitor water vapor concentration and temperature.

References:


Figure 4.1. Extinction coefficients at three wavelengths for water droplets at a temperature of 22°C with various size distribution functions, as shown in the inset. Refractive indices are taken to be $m=1.335$ at $\lambda=0.6328 \, \mu m$, $m=1.42-0.0195i$ at $\lambda=3.39 \, \mu m$, and $m=1.218-0.0508i$ at $\lambda=10 \, \mu m$.

Figure 4.1. Ratio of extinction coefficients between two wavelengths, $\lambda_1=1.5$ and $\lambda_2=0.5 \, \mu m$ for water droplets at 22°C with a log-normal size distribution function.
Figure 4.3. Schematic of differential absorption strategy for vapor measurement.

Figure 4.4. Extinction coefficients for water droplets with different diameters and vapor absorbance from water vapor from 0.5 to 9 µm at 22 °C.
Figure 4.5. $\Delta Q$ for water droplet with different diameters when $\lambda_3$ is selected at 5 $\mu$m.

Figure 4.6. Wavelength selection ($\lambda_3=2.6705$ and $\lambda_4=2.6818$ $\mu$m) of differential absorption scheme for water with a temperature of 22 $^\circ$C, total pressure 1 atm, mole fraction of water vapor 3%, and pathlength 1 cm.
Figure 4.7. Schematic used to evaluate the applicable range of differential absorption scheme for water vapor detection.

Figure 4.8. Comparison of droplet extinction and vapor absorption at a wavelength of $\lambda_3 = 2.6705 \ \mu m$ for the evaporation process depicted in Figure 4.7 to evaluate the applicable range of single wavelength scheme for vapor detection. Evaluation performed at a temperature of 22 °C, pressure 1 atm, and pathlength 1 cm.
Figure 4.9. Comparison of differential droplet extinction and vapor absorption between the wavelengths chosen in Figure 4.6 for the evaporation process depicted in Figure 4.7 to evaluate the applicable range of the differential absorption scheme for vapor detection. Evaluation performed at a temperature of 22 °C, pressure 1 atm, and pathlength 1 cm.

Figure 4.10. Schematic of multiphase diagnostic based on wavelength-multiplexed laser extinction.
Chapter 5

Experimental Demonstration of WMLE

5.1 Introduction

To test and validate the WMLE scheme developed in Chapter 3 and 4, a series of laboratory experiments have been conducted. This chapter describes three sets of experimental demonstrations of the WMLE scheme. In the first demonstration, a WJLE scheme was applied to solid polystyrene micro-spheres to demonstrate the concept. In the second example, a WMLE scheme was applied to measure the size distribution, mean diameter, and volume fraction in flows containing water aerosols generated by an ultrasonic nebulizer, and these measurements were compared against the measurements obtained using a Malvern Particle Sizer (MPS). In the third demonstration, a WMLE scheme was applied in an aerosol shock tube facility to monitor the interactions between shock waves and aerosol sprays. The WMLE scheme enabled simultaneous measurements of the aerosol size distribution, mean diameter, volume fraction, vapor concentration, and vapor temperature during such interactions with μs temporal resolution. Comparison between these measurements and shock wave calculations showed good agreement.
Chapter 5. Experimental Demonstration of WMLE

5.2 Demonstration 1: Polystyrene Micro-spheres

The first set of validation experiments was performed with polystyrene micro-spheres (Duke Scientific Corporation 7000 Series) because solid particles provide a highly controlled and repeatable sample for sizing experiments. In these experiments, polystyrene micro-spheres are suspended in water for measurement by an MPS (Malvern Instruments, Inc., Spraytec), a Coulter Multisizer (Coulter Electronics, Inc., model 0646), and a WMLE scheme consisting of four wavelengths (0.652, 1.33, 1.62, and 2.0 \( \mu \text{m} \)). These wavelengths are selected to avoid absorption by liquid water (the solvent) while maintaining good sizing accuracy and sensitivity following the analysis performed in Chapter 3. Note that in the demonstration here, the particle diameter does not vary in a range. Therefore, wavelengths in a wider spectral range as specified in Figure 3.9 is not necessary to achieve good measurement sensitivity. In the MPS and WMLE measurements, the micro-sphere solution is contained in a glass cell with 3 mm pathlength. Both MPS and Coulter Multisizer measurements suggest that log-normal distribution functions can be used to approximate the size distribution of the micro-spheres. Therefore, log-normal distribution functions are assumed in the WMLE measurements.

Figure 5.1 and Figure 5.2 show the comparison between size distribution measured by WMLE and MPS, by WMLE and Coulter Multisizer, respectively. Results in both figures show reasonable agreement between different size distribution measurement methods, validating the WMLE sizing technique and also indicating a log-normal distribution to be a good approximation for the micro-spheres. Note that MPS measures the volume distribution of particles, and the measured volume fraction needs to be divided by a factor of \( D^3 \) (\( D \) being the diameter of the particle) to obtain number distribution of the particles shown in Figure 5.1. Therefore, as illustrated in Figure 5.1, results from the MPS exhibit high uncertainty at small diameters. Measurements from the Coulter Multisizer also exhibit high uncertainty in the small diameter range, because the orifice of the testing tube (30 \( \mu \text{m} \) in diameter for the experiments shown in
Figure 5.2), while preventing impurities with diameter larger than 30 µm in the solution from entering the measurement, cannot prevent impurities with smaller diameter from entering the measurements.

Table 5.1 summarizes the measurement results of mean diameter (D_{mean}) and standard deviation (STD) shown in Figure 5.1 and Figure 5.2, and compares the results with manufacturer specifications. The mean diameter (D_{mean}) of droplets with a distribution function of f(D) is defined by the following equation:

$$D_{mean} = \int_0^\infty D \cdot f(D) \cdot dD$$  \hspace{1cm} (5.1)

For a log-normal distribution as defined in Equation (3.5), D_{mean} relates to \overline{D} and \sigma using in Chapters 3 and 4 by the following equation:

$$D_{mean} = \overline{D} + \exp\left[\frac{(\ln \sigma)^2}{2}\right]$$  \hspace{1cm} (5.2)

In Table 5.1, mean diameter and standard deviation (STD) are calculated from measured distributions of the micro-spheres and compared with the specifications provided by the manufacturer. From Table 5.1, we can see good agreement between different measurements and manufacturer specifications, which demonstrates and validates the WMLE sensing technique.

### 5.3 Demonstration 2: Ultrasonic Nebulizer

Ultrasonic nebulizers provide a very effective method to generate small aerosols by transferring acoustic energy to the nebulized liquid, and have attracted the attention of researchers from different disciplines [1-3]. The ultrasonic nebulizer is also an attractive device in the test and validation of the WMLE scheme due to the repeatability, spatial uniformity, and small diameter of the aerosols produced by ultrasonic nebulization. Here we developed a WMLE diagnostic utilizing seven wavelengths to simultaneously characterize both the aerosols and the vapor.
created by an ultrasonic nebulizer. Four of these wavelengths are used to measure the size distribution, mean diameter, and volume fraction of the aerosols, and the rest three are used to measure vapor concentration and vapor temperature.

The experimental arrangement for these measurements is shown in Figure 5.3. A commercially available ultrasonic nebulizer (Sonae Ultrasonics 241T) was operated at 2.4 MHz to atomize liquid water. Water aerosols produced were housed in a cylindrical chamber and convected into an optically-accessible test section by an air flow just above the liquid surface. The WMLE scheme and an MPS (Malvern Instruments, Inc., Spraytec) were applied at the test section (10 cm above the liquid surface) to measure the size distribution and volume fraction of the water aerosols. The WMLE sensor employs four wavelengths, 652 nm, 1.62 µm, 2.0 µm, and 3.39 µm, for aerosol size and distribution measurements. These wavelengths are selected based on the optimization procedures discussed in Chapter 3 and considering laser availability. Extinction by aerosols at these wavelengths was measured and used to infer mean diameter, volume fraction, and size distribution of the aerosols at the test station using the method described in Chapter 3. Note that in this application, the aerosol diameter does not vary in a range from 0.7 to 10 µm as analyzed in Chapter 3. Therefore, wavelengths in a narrower spectral range than that specified in Figure 3.9 can yield good measurement sensitivity. As discussed in Chapter 3, some \textit{a priori} information about the size distribution function needs to be assumed. Based on previous studies [4], we assumed that the diameter of the aerosols generated by the nebulizer follows a log-normal distribution as defined in Chapter 3. Another three wavelengths, 1.33, 1.382, and 1.390 µm, are incorporated in the WMLE for water vapor partial pressure and water vapor temperature measurements.

Example results of Sauter mean diameter ($D_{32}$) and volume fraction of the aerosols at the test station obtained by the WMLE sensor are shown in Figure 5.4 and Figure 5.5 respectively. In these measurements, a continuous air flow with a flow rate of 0.20 L/s was used to transport the
aerosols to the test section. As indicated in Figure 5.4 and Figure 5.5, aerosols arrived at the test station about 10 seconds after the nebulizer was turned on. Then both the Sauter mean diameter and volume fraction of the aerosols increased during a transient period, and became stable after about 40 seconds after the nebulizer was turned on. The stable Sauter mean diameter and volume fraction were about 2.75 µm and 26 ppm respectively in this experiment. Similar trends of Sauter mean diameter and volume fraction were obtained when other air flow rates were used.

The MPS was also applied to monitor the aerosols at the same location. Figure 5.6 shows the comparison between the WMLE measurements and the MPS measurements 60 seconds after the nebulizer was turned on at an air flow rate of 0.20 L/s. As mentioned above, a log-normal distribution was assumed for the diameter of aerosols in the WMLE measurements. The Comparison in Figure 5.6 shows good agreement between the WMLE and MPS measurements of the stable values of Sauter mean diameter and volume fraction, providing confirmation of the assumption that the diameter of the aerosols follows a log-normal distribution.

Figure 5.7 shows the water vapor partial pressure measured by the WMLE sensor, with comparison to hygrometer measurement and saturation water vapor pressure. As shown in Figure 5.7, prior to the arrival of water aerosols at the measurement locations, the WMLE reports a water vapor partial pressure of 10.8 torr, which is in good agreement with hygrometer measurements. About 10 seconds after the nebulizer is turned on, the evaporating water aerosols arrive at the test location and consequently the partial pressure of water vapor at the measurement location increases. After the transient period, water vapor partial pressure stabilizes at about 17.1 Torr, which is slightly lower than the vapor pressure of water at 21.3 °C, which was the ambient temperature when the experiment was performed. This is due to the fact that the temperature inside the test chamber is lower than ambient temperature due to evaporation of the water aerosols, which is experimentally confirmed by measuring the temperature inside the chamber using thermocouple. The temperature inside the chamber was measured to be 19.7 °C, which
corresponds to a water vapor pressure of 17.23 Torr. This is in good agreement with the WMLE measurement as shown in Figure 5.7.

5.4 Demonstration 3: Aerosol Shock Tube

5.4.1 Experimental Method

Shock tubes offer a number of advantages for studying the interactions of shock waves and aerosols, and laser diagnostics developed for these studies show promise as a powerful tool [5-8]. However, most past work relied on a few wavelengths in a narrow spectral range (usually from the visible to near-infrared range) for the monitoring of aerosols. In this section, we describe the application of a WMLE diagnostic consisting of eight wavelengths in a wider spectral range in an aerosol shock tube facility for the simultaneous characterization of aerosols and vapor.

The schematic of the diagnostic applied to the shock tube is illustrated in Figure 5.8. The shock tube used in these experiments has been discussed in detail in earlier papers [4, 9]. The operation of this shock tube is briefly reviewed here for convenience. First, the driver and driven sections of the shock tube are evacuated and then filled with argon to the desired pressure. Second, water aerosols generated by an ultrasonic nebulizer are fed into the shock tube. Finally, the pressure in the driver section is raised by filling more argon to rupture the diaphragm and form a shock wave. The shock wave develops and propagates along the tube, and interacts with the aerosols in the driven section. These interactions are monitored by the WMLE sensor located near the end wall of the tube as shown in Figure 5.8.

The WMLE sensor employs seven wavelengths ranging from 0.652 to 10.0 μm for simultaneous measurement of size distribution and volume fraction of the aerosols, and concentration and temperature of water vapor. Absorption by water vapor does not occur at the
following wavelengths, 0.652, 1.33, 1.62, 1.997, 3.39, and 10.0 μm. Extinction at these wavelengths is due to aerosol scattering. Therefore, size distribution and volume fraction of the aerosols are extracted from the extinction measurements at these wavelengths using the method developed in Chapter 3. More specifically, accuracy and sensitivity of diameter and distribution measurement (as defined in Equations 3.9 and 3.10) by all the possible wavelength combinations among these six wavelengths are evaluated. Different wavelength combinations are applied in different in different ranges of aerosol diameter to achieve maximum measurement accuracy and sensitivity. In these experiments, a log-normal distribution function as defined in Equation 3.5 is again assumed for the diameter of the aerosols. Furthermore, as revealed by previous characterization of the nebulizer, \( D_{32} \) and \( \sigma \) of the aerosols generated by the nebulizer are around 3 μm and 1.4 respectively. According to the evaluation, for such log-normal distributions, the wavelength combinations shown in Figure 5.9 yield the maximum measurement accuracy and sensitivity. Note that due to the availability of the laser sources when these experiments were performed, the wavelengths selected for aerosol measurement are not fully optimized. Therefore as indicated in Figure 5.9, the performance of this WMLE scheme does not reach that shown in Figure 3.9. For example, the sensitivities of \( D_{32} \) and \( \sigma \) measurements is lower, and the ratio of extinction (R) is less than 0.1 for the wavelength combination of 10 and 0.6328 μm in part of its applicable \( D_{32} \) ranges. Incorporation of a short wavelength (e.g. 244 or 266 nm) will enhance the measurement performance and extend the measurable \( D_{32} \) to wider ranges.

Concentration and temperature of water vapor are extracted from the extinction measurements at wavelengths of 1.33, 1.3899, and 1.3917 μm. As indicated by the absorption spectra of water vapor near these wavelengths (simulated by HITRAN 2004) shown in Figure 5.10, water vapor absorbs the laser radiation at 1.3899 and 1.3917 μm, but not at 1.33 μm. More specifically, the absorption strength of the absorption feature centered at 1.3899 μm decreases with increasing temperature, and that of the absorption feature centered at 1.3917 μm increases
with increasing temperature. The differences in extinction measurements between 1.33 µm and
the other two wavelengths (1.3898 and 1.3917 µm) as described in Chapter 4 are calculated to
infer the absorbance due to water vapor at these wavelengths. At a given pressure, the ratio of
absorbance at these two wavelengths only depends on temperature and therefore is used to infer
vapor temperature. After temperature is measured, absorbance at either wavelength (1.3898 or
1.3917 µm) can be used to infer concentration of water vapor. References [10, 11] provide more
information about the gas temperature measurement using two-wavelength absorption. Pressure is
measured by piezoelectric pressure transducers (PCB Piezotronics, model 13A26) in these
experiments. Laser intensity at each wavelength and pressure signal are recorded at 10 MHz
using a digital oscilloscope.

### 5.4.2 Aerosol Measurements

Figure 5.11 and Figure 5.12 show the results of the size distribution and Sauter mean
diameter measurements in a typical shock wave experiment with argon as background gas. For
these measurements, the pre-shock pressure and temperature are \( P_1 = 0.263 \text{ atm} \) and \( T_1 = 293 \text{ K} \),
respectively. Incident and reflected shock conditions calculated considering only the vapor
components are: \( T_2 = 510 \text{ K} \), \( P_2 = 0.965 \text{ atm} \), \( \rho_{21} = 2.12 \), \( T_5 = 769 \text{ K} \), \( P_5 = 2.644 \text{ atm} \), and \( \rho_{52} = 1.82 \).
Shock attenuation in these aerosol experiments is about 3% per meter. In the experimental
conditions specified above, the increase in \( T_2 \) due to shock attenuation is less than 10 K in the 500
µs measurement time after the incident shock. In this experiments, the pre-shock size distribution
of the aerosols is determined to be a log-normal distribution with \( D_{32} = 2.65 \text{ µm} \) and \( \sigma = 1.45 \) as
shown in Figure 5.11. According to [12], these calculations provide a close approximation for
pressure and temperature immediately behind the incident shock front. From Figure 5.12, we see
that the pre-shock Sauter mean diameter of the aerosols is 2.65 µm, which agrees with the
measurement using the MPS. At time zero, the incident shock wave passes through the laser beam and deflects the laser beam from the detectors, which results in a sharp spike in the measurements as shown in Figure 5.12. A similar feature occurs upon the arrival of the reflected shock wave. After shock arrival, the mean diameter of the aerosols decreases due to evaporation brought about by shock heating, which is captured by the measurement shown in Figure 5.12.

Note that in Figure 5.12, the time scale between the incident and reflected shock has been converted to particle time by multiplying the laboratory values by $\rho_21$. The same conversion is performed for all the subsequent figures.

Figure 5.13 shows the results of aerosol number density measurement in the same shock wave experiment. Note that once the size distribution of the aerosols is determined, as shown in Figure 5.12, extinction from any of the wavelengths (0.652, 1.33, 1.62, 1.997, 3.39, and 10.0 $\mu$m) can yield the number density information. In Figure 5.13, the number densities of the aerosols inferred using extinction measurements at 1.33 and 1.62 $\mu$m are shown and are in agreement as expected. The number density remains constant after arrival of the incident and reflected shock wave, as shown in Figure 5.13, indicating that no significant coalescence or breakup of the aerosols occur during their interactions with the shock wave. The changes in aerosol number densities across the shock wave are due to shock compression. Figure 5.13 also shows the number density of the aerosols calculated based on shock calculations using only the vapor components. In these calculations, the pre-shock number density ($n_1$) is considered to increase by a factor of $\rho_21$ across the incident shock wave due to shock compression. The close agreement between this calculated value and the measured value of $n_2$ supports the approximation of shock calculations immediately after the arrival of incident shock wave by using only the vapor components. However, evaporation of the aerosols afterwards results in temperature changes of the bulk flow. Therefore, prediction of the number density after the reflect shock wave ($n_3$) neglecting aerosols in the flow significantly deviates from the measured value, as shown in Figure 5.13.
Figure 5.14 shows the measured volume fraction of the aerosols in this shock wave experiment. It can be seen that the pre-shock volume fraction of the aerosols is 7.0 ppm. Similar to the above discussion about number density measurement, the volume fraction of the aerosols can also be estimated based on shock calculations using only the vapor components. Figure 5.14 illustrates the comparison between such calculations and the measured value. Figure 5.14 shows the evaporation of water aerosols due to shock heating, resulting in decreasing volume fraction of the aerosols after each shock arrival.

Finally, these measurements of the aerosols enable the inference of an evaporation rate constant. The pre-shock aerosols (with measured size distribution and initial volume fraction) are expected to evaporate according to a $D^2$-Law (as shown in the following equation) after shock arrival:

$$D_0^2 - D(t)^2 = kt$$

(5.3)

In Equation (5.3), $t$ is time after evaporation occurs, $D_0$ the initial droplet diameter at $t=0$, $D(t)$ the droplet diameter at time $t$, and $k$ the evaporation rate constant. Equation (5.3) describes the diminishment of the droplet diameter undergoing steady-state evaporation.

The time history of such evaporation is reflected in the volume fraction measurement as shown in Figure 5.14. For a given set of pre-shock conditions of the aerosols (size distribution and initial volume fraction), the evolution of the volume fraction is determined by the evaporation rate constant. In this work, we developed a model based on the $D^2$-Law evaporation to calculate the evolution of the volume fraction, and used this model to infer the evaporation rate constant that results in the best fit between the calculated and measured volume fraction. More specifically, the inference of evaporation rate constant by this model includes the following five steps. First, the pre-shock number density and size distribution function of the aerosols are calculated from extinction measurements at wavelengths in the WMLE sensor, as shown in Figure 5.11 and Figure 5.12. The number density and size distribution function of the aerosols immediately after
incident shock arrival are assumed to be the same as the pre-shock values. Therefore, this step essentially provides the initial conditions for the model by assuming that 1) no aerosol coalescence or breakup occurs during the incident shock arrival, which is confirmed by the measurements shown in Figure 5.11, and 2) no evaporation of the aerosols occurs during the incident shock arrival, which is confirmed by the agreement between volume fraction measured and calculated based on shock calculations as shown in Figure 5.12. Second, the evolution of the distribution function under the D²-Law evaporation as defined in Equation (5.1) is calculated by discretizing the distribution function and applying an assumed evaporation rate constant. Third, the time history of the volume fraction of the aerosols is obtained by combining the distribution function calculated in the second step and the number density. Fourth, different evaporation rate constants are tried in the second step to calculate the evolution of the distribution function, which is then used as input in the third step to calculate the evolution of the volume fraction. Fifth, the calculated time histories of the aerosol volume fraction are compared with the measured one to find the best least-square fit between the calculation and the measurement. The evaporation rate constant which provides the best fit is determined to be the evaporation rate constant.

For example, when applied to the above shock wave experiment, this model determines an evaporation rate constant of 0.020 mm²/s, which provides the best fit of the measured volume fraction data after the incident shock wave arrival and prior to the reflected shock arrival, as shown in Figure 5.14. From shock calculations listed above, the bulk gas temperature immediately after incident shock arrival is 510 K. Thus, the evaporation rate constant of water at a temperature of 510 K is determined to be 0.020 mm²/s. Figure 5.14 also shows the history of droplet volume fraction at other two evaporation rate constant, 0.017 and 0.023 mm²/s to illustrate the sensitivity of data fitting. The evaporation rate constants at other temperatures can be determined similarly. Evaporation rate constants determined in this work are in good agreement with evaporation rate constant inferred from a different approach [9]. In [9], Hanson et al. developed a code to compute the Mie extinction by aerosols following log-normal distributions
modified by D²-Law evaporation as defined in Equation (5.1). The calculated Mie extinction depends on the initial aerosol size distribution and the modification by D²-Law evaporation, which depends on evaporation rate constant. Then the evaporation rate constant is determined by finding the best fit between the calculated Mie extinction and the measured Mie extinction at wavelengths employed in the WMLE sensor. Evaporation rate constant of water at a temperature of 510 K measured in [9] is 0.023 mm²/s. Refer to [9] for more discussion about the experiments to measure evaporation rate constants in this aerosol shock tube facility.

The evaporation rate constant can also be calculated from theoretical model of droplet evaporation. The evaporation rate constant for water at a temperature of 510 K is calculated to be 0.019 mm²/s, which is in reasonable agreement with the value determined here. Detailed calculation of the evaporation rate constant can be found in Appendix D.

Note that the bulk gas temperature decreases during the time period in consideration due to evaporation of the aerosols, as shown by vapor temperature measurements in the next section. Therefore, it is difficult to assign the evaporation rate constant to a single temperature. Nevertheless, these aerosol measurements provide an effective way to determine the evaporation rate constants, and ambiguity due to uncertainty in temperature can be reduced by developing better models to incorporate vapor temperature measurements and consider the effect of varying temperature.

The measurement location in these experiments is 10 cm away from the end wall. Shock wave calculations at such distance are not reliable in the reflected shock region, therefore preventing meaningful comparison between WMLE measurements and shock wave calculations in the reflected shock region.
5.4.3 Vapor Measurements

Figure 5.15 shows the corresponding water vapor temperature measurements for the shock tube experiment specified in the proceeding section. The pre-shock temperature measured by the WMLE sensor is 293 K, which agrees with temperature measured by thermocouples. From Figure 5.15, we can see that the water vapor temperature immediately behind the incident shock passages approximates that from shock calculations neglecting aerosols. During evaporation of the aerosols in the shock heated gas, the bulk gas temperature decreases as expected. After water temperature is determined, partial pressure (or mole fraction) of water vapor can be determined using extinction measurement at either 1.3899 or 1.3917 µm combined with pressure measurement. Figure 5.16 shows the partial pressure of water vapor determined using the extinction measured at 1.3917 µm because of the stronger absorption strength at this wavelength at temperatures near 510 K (T_2). Again, the partial pressure of water vapor immediately behind the incident shock passages can be approximated by shock calculations as suggested by Figure 5.16. The partial pressure of water increases during evaporation, as shown in Figure 5.16.

Obviously, the aerosol and vapor measurements for a same shock wave experiment should be related. In this work, we attempted to reveal such relationship by modeling the water vapor temperature based on the measure volume fraction of the aerosols and comparing the model results to the measured temperature profile, as shown in Figure 5.15. We assumed a temperature of 510 K (T_2) for the bulk gas immediately after the incident shock arrival, and calculated the subsequent bulk gas temperature changes due to evaporation of the water aerosols based on the volume fraction measurements shown in Figure 5.14. The energy transfer from the hot gas to the aerosols during the evaporation process is modeled as a closed system at constant pressure. The following conservation equations of energy and mass explain the calculation of the bulk gas temperature in more details:

\[ m_{av} \cdot H_{av}(T_0) + m_{v} \cdot H_{v}(T_0) + m_{l} \cdot H_{l}(T_L) = m_{av} \cdot H_{av}(T) + m_{v} \cdot H_{v}(T) + m_{l} \cdot H_{l}(T_L) \]  (5.4)
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\[ m_{Ar} + m_{v}^0 + m_{L}^0 = m_{Ar} + m_{v} + m_{L} \]  \hspace{1cm} (5.5)

where:

- \( m_{Ar} \) = mass of argon vapor in the closed system
- \( H_{Ar}(T_0) \) = enthalpy of argon at a temperature of \( T_0 \) (time = 0)
- \( T_0 \) = bulk gas temperature immediately after the incident shock wave (time = 0)
- \( m_{v}^0 \) = mass of water vapor immediately after the incident shock wave (time = 0)
- \( H_{v}(T_0) \) = enthalpy of water vapor at a temperature of \( T_0 \) (time = 0)
- \( m_{L}^0 \) = mass of liquid water immediately after the incident shock wave (time = 0)
- \( H_{L}(T_L) \) = enthalpy of liquid water at a temperature of \( T_L \) (time = 0)
- \( T_L \) = temperature of the liquid water aerosols (approximated by the surface temperature of the aerosols, which is calculated by the method described in Appendix D)
- \( H_{Ar}(T) \) = enthalpy of argon at a temperature of \( T \) (time = t)
- \( m_v \) = mass of water vapor at time t
- \( H_{v}(T) \) = enthalpy of water vapor at a temperature of \( T \) (time = t)
- \( m_L \) = mass of liquid water at time = t

The definition of the volume fraction \((C_v)\) is shown below

\[ C_v = \frac{V_L}{V_L + V_v} \approx \frac{V_L}{V_v} = \frac{m_L/\rho_L}{m_{Ar}/\rho_{Ar} + m_v/\rho_v} \]  \hspace{1cm} (5.6)

where:

- \( V_L \) = volume of liquid (in the form of aerosols) in the system at time t
- \( V_v \) = volume of vapor in the system at time t
- \( \rho_{Ar} \) = density of the vapor at time t (temperature T)
- \( \rho_L \) = density of the liquid (in the form of aerosols) at time t (temperature T)
In the derivation of Equation (5.3), the fact that \( V_L << V_V \) is used to simplify the denominator of the definition. Three unknowns exist in the above three equations, gas temperature at time \( t \) (\( T \)), mass of water vapor and liquid water at time \( t \) (\( m_V \) and \( m_L \)). Therefore, enclosure is obtained within these three equations. An iteration solution to the system of Equations 5.4, 5.5 and 5.6 provides gas temperature and mass fraction of water vapor at time \( t \).

Figure 5.15 and Figure 5.16 show the solution of gas temperature (\( T \)) and water vapor partial pressure (\( P_V \)) from the model, using the measured \( C_V \) as input in Equation 5.6. Note that mass of water vapor solved from Equations 5.4, 5.5 and 5.6 relates to partial pressure (\( P_V \)) by the following equations:

\[
P_V = \chi_V \cdot P \\
\chi_V = \frac{m_v}{M_v} \cdot \frac{1}{m_v/M_v + m_{Ar}/M_{Ar}}
\]

where:

- \( \chi_V \) = mole fraction of water vapor in the mixture
- \( P \) = total pressure of the mixture, measured by piezoelectric pressure transducers
- \( M_V, M_{Ar} \) = molecular weights of water and argon, respectively

As illustrated in Figure 5.15 and Figure 5.16, the model result approximates the magnitude and trend of the measured temperature and partial pressure profile, though some discrepancies exist. These discrepancies in this comparison can be better resolved by 1) developing a more complete model (such as considering heat loss during the shock-aerosol interactions in the model) to predict the temperature of water vapor, 2) refining our WMLE measurements (such as quantifying the refractive index of the aerosols at elevated temperatures) to provide improved experimental data, and 3) considering the nonuniform temperature distribution around the aerosols both in the measurement and in the model.
Similar to the discussion in Section 5.3.3, shock wave calculations at the measurement location are not reliable in the reflected shock region, therefore preventing meaningful comparison between experimental data and shock wave calculations in the reflected shock region.

5.5 Summary

Laser diagnostics based on the WMLE concept have been developed and demonstrated in a series of experiments. In experiments using an ultrasonic nebulizer and an aerosol shock tube facility, the WMLE technique enabled simultaneous measurements of aerosol size distribution, aerosol volume fraction, vapor temperature, and vapor concentration with µs time response. These encouraging preliminary results demonstrate the feasibility and utility of the WMLE technique. This diagnostic should provide a powerful tool to advance studies of heterogeneous flows, including spray evaporation, droplet combustion, detonation of sprays, and combustion quenching.

References:


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Table 5.1 Comparison of micro-sphere size distribution measurement by WMLE, MPS, and Coulter Multisizer

<table>
<thead>
<tr>
<th></th>
<th>WMLE</th>
<th>MPS</th>
<th>Coulter Multisizer</th>
<th>Manufacture’s Specification</th>
</tr>
</thead>
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<tr>
<td>$D_{\text{mean}}$ (µm)</td>
<td>4.18</td>
<td>3.97</td>
<td>4.12</td>
<td>4.3</td>
</tr>
<tr>
<td>STD/$D_{\text{mean}}$</td>
<td>24.7%</td>
<td>26.3%</td>
<td>23.1%</td>
<td>≤ 25%</td>
</tr>
</tbody>
</table>

Figure 5.1. Comparison of droplet size distribution measurement by MPS and WMLE in microsphere solution.
Figure 5.2. Comparison of droplet size distribution measurement by Coulter Multisizer and WMLE in micro-sphere solution.

Figure 5.3. Schematic diagram of WMLE sensor applied to an ultrasonic nebulizer.
Figure 5.4. Sauter mean diameter measured by the WMLE sensor.

Figure 5.5. Volume fraction measured by the WMLE sensor.
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Figure 5.6 Size distribution measured by the WMLE sensor, with comparison to the measurements by an MPS.

Figure 5.7. Water vapor partial pressure measured by WMLE sensor, with comparison to hygrometer measurement and saturation water vapor pressure.
Figure 5.8. Experimental arrangement of the WMLE sensor applied to the aerosol shock tube facility.

Figure 5.9 Summary of the applicable ranges of different wavelength combinations in the WMLE scheme shown in Figure 5.8 for aerosol measurements.
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Figure 5.10. Absorption spectra of water vapor simulated by HITRAN 2004 near the wavelengths used in the WMLE sensor for vapor measurement.

Figure 5.11. Pre-shock aerosol size distribution function.
Figure 5.12. Sauter mean diameter of the aerosols measured by the WMLE sensor.

Figure 5.13. Number density of the aerosols inferred from extinction measurements at different wavelengths in the WMLE sensor.
Figure 5.14. Volume fraction of the aerosols measured by the WMLE sensor, with comparison to calculation with an evaporation rate constant of 0.020 mm²/s.

Figure 5.15. Water vapor temperature measured by WMLE sensor after shock wave passage with comparison to shock calculations.
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Figure 5.16. Partial pressure of water vapor measured by WMLE sensor after shock wave passage with comparison to shock calculations.
Chapter 6

Summary and Future Work

6.1 Summary

Laser diagnostics based on extinction measurement at multiplexed wavelengths have been developed for the simultaneous characterization of vapor and droplets in sprays. This work first developed the sensing technology for vapor sensing using spectrally resolved absorption spectroscopy, with application in practical engines for fuel monitoring and active engine control. The vapor measurement method was then extended to the WMLE concept for the simultaneous measurement of multiple parameters in sprays by exploiting wavelength multiplexing in a broad spectral range. Diagnostics based on the WMLE concept were developed and demonstrated with polystyrene micro-spheres, then in cold-flow spray systems, and finally in an aerosol shock tube to simultaneously measure aerosol and vapor properties during interactions between shock waves and the aerosols.

Key virtues of the WMLE technique demonstrated in these applications are that 1) it offers good prospects for near-real-time monitoring capability (1 µs for droplet sizing and vapor sensing was demonstrated in the aerosol shock tube applications), and 2) it extends the capability of near-IR diode laser gas sensors previously developed to multiphase flows. These successful
demonstrations provide strong support for the further development and expanded applications of the WMLE technique.

### 6.2 Future Work

#### 6.2.1 Further Applications of the C$_2$H$_4$ Sensor

The method of integrating spectral absorbance to infer C$_2$H$_4$ concentration as described in Chapter 2 has the advantages of being accurate and of utilizing all the spectral information available, but requires intense computation and is therefore not practical for real-time control applications. In the control applications described in Chapter 2, the detector signal is processed by a lock-in amplifier synchronized with the diode laser wavelength scan rate to provide qualitative information of C$_2$H$_4$ concentration. With an expanded spectroscopic database of C$_2$H$_4$, e.g. as shown in Figure 2.8, some simplified algorithms (e.g., inference of C$_2$H$_4$ concentration based on the absorbance at a few frequencies within the scanned spectra) can yield quantitative measurement of C$_2$H$_4$ concentration with temporal resolution of 0.2 ms. With a faster algorithm, this sensor shows promise of enabling a more quantitative scheme for active control of C$_2$H$_4$-fuled combustion systems. The expanded spectroscopic database also extends the utility of the sensor from atmospheric pressure to other pressures, and allows simultaneous measurements of multiple parameters. Finally, the temporal resolution of the sensor in this work is 0.2 ms. Better temporal resolution can be readily achieved by scanning the laser at a faster rate to measure the spectra of C$_2$H$_4$.

#### 6.2.2 Further Development of the WMLE Sensor

One important aspect of the further development of the WMLE technique is the extension of the WMLE schemes described in this work to simultaneous vapor and droplets measurements.
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in hydrocarbon fuel sprays. The extension of WMLE technique to hydrocarbon fuels requires research work to address the following two key topics.

First, studies of gas phase spectra of target fuel species need to be conducted, including the temperature dependence of the vapor spectra. A good understanding of the vapor spectra is necessary to optimize wavelengths for differential absorption, to enable quantitative vapor concentration measurements, and to obtain simultaneous vapor temperature measurements. The recent availability of tunable mid-IR laser sources has the potential to significantly facilitate the sensing of hydrocarbon fuel vapor based on differential absorption. These laser sources offer new opportunities to utilize the strong absorption by most hydrocarbon vapors near 3.3 \( \mu \text{m} \) and to discriminate against interference droplet extinction due to their wavelength tunability.

Second, liquid phase absorption spectra of target fuels need to be determined. As discussed in Chapter 3, both the design of the WMLE technique and the interpretation of the measurements depend on the refractive index of the target liquid, which is derived from liquid phase spectra. Therefore, a reliable liquid spectra database lays the groundwork for droplet sizing by the WMLE technique. A quantitative understanding of the relationship between refractive index and temperature of the target liquids also merits research effort. This understanding will facilitate the quantitative application of the WMLE scheme at elevated temperatures, and also has the potential to enable non-intrusive measurements of aerosol temperature when the size distribution function of the aerosols is measured by an independent method or by an independent set of wavelengths in the WMLE sensor.

Other aspects of the further development of the WMLE technique include the concept of dense-wavelength-multiplexing for measurement of more complicated distribution functions (for example, multi-modal distributions) and the extension of the WMLE technique to achieve spatially resolved measurements to deal with no uniformity along the pathlength. WMLE schemes are considered mainly for distribution functions with two parameters or three parameters in this work. More wavelengths will be required for the measurement of more complicated
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distribution functions. Research effort will be needed to optimize laser wavelengths and the number of detection channels, as a function of droplet size and distribution shape, for optimized sensor performance. Development and application of the WMLE scheme in this work focused on line-of-sight measurements. Extensions of these concepts to allow spatially resolved measurements can be envisaged by incorporating additional wavelengths and measuring the Mie-scattering at right angles, and by splitting the probe beam to measure extinction at multiple locations simultaneously to provide a tomography of the vapor and droplet properties.

With further research and development, the WMLE technique promises to be a powerful tool for fundamental and applied studies of heterogenous flows where highly transient phenomena, such as spray mixing, evaporation, and combustion are important.
Appendix A
Survey of Atmospheric Aerosol Measurements

There is such a large body of literature existing on atmospheric aerosol measurements distributed in very diversified communities that it would appear irrelevant to review this topic in the thesis. This appendix attempts to provide a brief survey of the techniques for atmospheric aerosol measurements, with emphasis on contrasting the different measurement needs in this thesis and in general atmospheric aerosol measurements.

Atmospheric aerosol measurements represent a large body of science of great importance to many different research communities, ranging from pollutant monitoring, climate change, health effects, and formulation of national ambient air quality standard. Consequently, numerous methods for atmospheric aerosol measurements have been developed during the past four decades. Refer to [1-5] for a complete review of these measurement techniques used for atmospheric monitoring applications. This appendix will discuss the special measurement needs and requirements in current fuel aerosol work in contrast with published atmospheric aerosol measurements.

Atmospheric aerosol measurements in general are more complicated than the measurements described in this work. First, atmospheric aerosols range in size over more than five orders of magnitude, from nuclei with diameters of a few nanometers to cloud droplets up to tens of microns. No single technique can provide satisfactory measurement over this entire size range of interest. Therefore, an array of different techniques has been developed for measurement of aerosol properties in different size ranges. These techniques include condensation particle counters for the measurement of number density of aerosols with
diameter down to 0.003 µm [6, 7], filtration gravimetric for the measurement of mass fraction of aerosols with diameter above 0.1 µm [3], single-particle optical counters for size measurement of aerosol with diameter above 0.05 µm [8]. In contrast, a relatively narrow size range (~1 to 10 µm) is of interest to this current work, and consequently a single technique (wavelength-multiplexed laser extinction) can provide satisfactory measurement of multiple aerosol properties (size, number density, volume fraction, etc.) in this entire size range. Second, chemical composition of atmospheric aerosols is extremely complicated. Though water typically constitutes more than half of the atmospheric aerosol mass (depending on ambient humidity), the rest of aerosol mass contains a great number of species such as salt, soot, and metals, nitrates, and hundreds of organic compounds. Most aerosol measurement techniques depend on the chemical composition of the aerosols, therefore, measurement of the chemical composition itself is an important topic in atmospheric aerosol measurement. Measurement of chemical composition of aerosols typically involves off-line chemical analysis of the collected aerosol samples [9, 10]. Real-time measurement of chemical composition of atmospheric aerosols is still under development [11, 12]. In contrast, aerosols considered in this thesis are of single chemical composition or of well-defined composition, which renders it possible for the real-time measurement based on optical scattering methods. Lastly, the effect due to irregular aerosol shapes must be considered in atmospheric aerosol measurement [13]. Although liquid aerosols can be well approximated as spheres, atmospheric aerosols usually contain a mixture of liquid spheres, irregularly shaped solid particles, and solid particles encapsulated by liquids. Only homogeneous liquid aerosols are of interest to the applications in this work, which substantially simplifies the optical measurement technique because light scattering can be rigorously derived from Mie scattering theory in this case.

In spite of all the simplicities listed above, the current work has some measurement requirements distinct from those for atmospheric aerosol measurements. First, non-intrusive
measurement is highly desirable, if not required, in most propulsion applications. Optical measurement is so far the best solution for non-intrusive measurements. Second, applications in this work require *in-situ* measurement with high temporal response. In the experiments performed in the aerosol shock tube described in Chapter 5, a temporal response on the order of 1 µs or better is required. Other propulsion applications (e.g., applications in pulse detonation engines operating at high repetition rate) require similar temporal response. Among all the optical methods investigated in this work, extinction measurement offers the best promise to achieve such temporal resolution. Third, in most propulsion applications, vapor properties are of equal importance to droplet properties. Extinction and absorption based on wavelength-multiplex laser systems provide good perspective for the simultaneous monitoring of multiple aerosol and vapor properties.

In summary, though numerous methods have been developed for atmospheric aerosol measurements, wavelength-multiplexed laser extinction is well-suited to meet the special measurement requirements encountered in the applications described in this work.

References:

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Appendix B

Evaluation of Interference and Multiple Scattering in the WMLE Scheme

As mentioned in Chapter 3, all the discussions in this thesis have neglected the effects of interference scattering and have been based on the assumption of single scattering events. This appendix provides an evaluation of the influence of interference scattering in the WMLE scheme and a discussion on the applicable range of the single scattering event assumption.

As illustrated in Figure B.1, when a particle is illuminated by light with an intensity of $I_0$, energy intercepted (i.e. extinction of energy) by the particle is given by the following equation:

$$E_{ext} = \frac{\pi D^2}{4} \cdot Q_{ext} \cdot I_0$$  \hspace{1cm} (B.1)

in which:

- $E_{ext}$: extinction of energy by the droplet (W)
- $D$: diameter of the droplet ($\mu$m)
- $Q_{ext}$: extinction coefficient
- $I_0$: Intensity of incident light (W/m$^2$)

Detectors are used to register the transmitted light intensity, which is then used to inferred extinction. However, in practice, detectors are always of finite size (denoted by $L$ in Figure B.1), which translates to a finite acceptance angle (denoted by $\theta_{acc}$ in Figure B.1). Therefore, light intensity recorded by the detector contains both transmitted light and light scattered by the particle. Light scattered from the particle and collected by the detector poses...
an interference to the measurement of transmitted light, and consequently to the inference of extinction. Here, interference scattering resulting from the finite detection size (acceptance angle) is evaluated.

Energy scattered by the particle and collected by the detector is given by the following equation [1]:

\[
E_{\text{sca}} = 2\pi I_0 \int_{\theta=\theta_{\text{acc}}}^{\theta=\theta_{\text{acc}}} S(\theta) \cdot \sin(\theta) \cdot d\theta
\]

(B.2)

where:

\(E_{\text{sca}}:\) energy scattered by the particle and collected by the detector (W)

\(S(\theta):\) total phase function of scattering (m\(^2\))

In Equation (B.2), the total phase function, \(S(\theta)\), represents the fraction of incident energy scattered into unit solid angle about a direction which makes an angle \(\theta\) with the forward beam. Figure B.2 shows an example calculation of \(S(\theta)\) normalized by \(S(\theta=0^\circ)\) at a wavelength of 0.6 \(\mu\)m and droplet diameter 4 \(\mu\)m. From the inset of Figure B.2, it can be seen that almost all the scattered light is confined within a cone of half-angle of 10\(^0\). Therefore, \(\theta_{\text{acc}}\) must be much smaller than this angle to record the true extinction signal. In general, almost all the scattered light is confined within a cone of half-angle of \(10\lambda/\pi D\) [1]. Therefore, for droplets with fixed diameter, the shortest wavelength in the WMLE system defines the maximum \(\theta_{\text{acc}}\). Here, a wavelength of \(\lambda=0.6\) \(\mu\)m is used to evaluate the interference scattering.

In this work, the ratio between \(E_{\text{sca}}\) and \(E_{\text{ext}}\) as defined below is used to quantify the interference of spurious scattering to the extinction measurement:

\[
R = \frac{E_{\text{sca}}}{E_{\text{ext}}} = \frac{2\pi I_0 \int_{\theta=\theta_{\text{acc}}}^{\theta=\theta_{\text{acc}}} S(\theta) \cdot \sin(\theta) \cdot d\theta}{\pi D^2/4 \cdot Q_{\text{ext}} \cdot I_0} = \frac{8\int_{\theta=\theta_{\text{acc}}}^{\theta=\theta_{\text{acc}}} S(\theta) \cdot \sin(\theta) \cdot d\theta}{D^2 \cdot Q_{\text{ext}}}
\]

(B.3)

According to Equation (B.3), a large \(R\) means that interference scattering constitutes a significant part in the signal measured by the detector, and vice versa.

Figure B.3 shows the calculation of \(R\) for an acceptance angle in the range from 0 to
at a wavelength of 0.6 µm for water aerosols. From the inset of Figure B.3, we can see that an R of 1% corresponds to an acceptance angle of 0.78 °. Therefore, if one desires to limit the fraction of interference scattering in the extinction measurement below 1%, one must design the detection optics such that acceptant angle is smaller than 0.78 °. If the distance from the detector to the particle (denoted as R in Figure B.1) is 30 cm, which is the distance encountered in the experiments performed in Chapter 5, the radius of the active area of the detectors (or the radius of any mirror or focusing lens in the detection system) needs to be smaller than 4.0 mm. In the these experiments, iris diaphragms are used to reduce the acceptance angle to ~0.3 ° and ensure interference scattering has a negligible impact.

Similar analysis can be performed in the case when a collection of particles is in the pathlength instead of a single particle, and the results will be the same as in the single particle case as long as multiple scattering does not occur.

It is rather difficult to give general and definitive criteria under which the assumption of single scattering is valid [2]. See [2-5] for more detailed discussions regarding this topic. Here we limit our discussions to the conditions under which the WMLE scheme developed in Chapter 3 is applicable. Two conditions need to be considered [3, 4], which are described by the following tow equations:

\[
0 < \tau < 10 \quad \text{(B.4)} \\
\frac{d}{D} \ll 1 \quad \text{(B.5)}
\]

in which:

\( \tau \) = extinction by the aerosols as defined in Equation (3.1).

D = diameter of the aerosol

d = distance between aerosols

These two conditions are always satisfied in the experiments described in Chapter 5. Under typical experimental conditions, values of \( \tau \) and \( d/D \) are below 1.5 and greater than 50,
respectively. Therefore, application of the WMLE scheme with a single scattering event assumption is justified for these experiments.

References:


Figure B.1 Schematic illustrating the interference signal due to scattering registered by the detector.

Figure B.2 Normalized total phase function (normalized by $S(\theta=0)$) at $\lambda=0.6 \, \mu m$ and $D=4 \, \mu m$ for water droplet.
Figure B.3 Evaluation of interference scattering at different acceptance angles.
Appendix C
Influence of Aerosol Temperature on WMLE

The consideration of the influence of aerosol temperature on the WMLE technique essentially derives from the variation of the refractive index of the aerosols with temperature. When the aerosol temperature varies, refractive index of the aerosols varies, which will cause changes in the extinction coefficients and finally result in variations in the reference of droplet properties (mean diameter, size distribution, and volume fraction). In this appendix, the influence of aerosol temperature on the WMLE technique will be examined, and this appendix starts by investigating the dependence of extinction coefficients on refractive index of the target aerosols.

Figure C.1 shows some example calculations to illustrate the dependence of extinction coefficients on refractive index of the aerosols. First, the extinction coefficient of water aerosol at a wavelength of 2 μm is calculated based on a refractive index of m=1.306-0.0011·i (the refractive index of water at 2 μm at a temperature of 22 °C). Then extinction coefficients are calculated at other modified refractive indices. Figure C.1 shows that a 25% decrease in the imaginary part of the refractive index has negligible impact on extinction coefficients. Actually, the extinction coefficient calculated at m=n-1.25k·i virtually overlaps with that calculated at m=n-k·i as shown in Figure C.1. Even varying the imaginary part by one order of magnitude does not change the extinction coefficient significantly. This observation, i.e., extinction coefficients have a weak dependence on the imaginary part of the refractive index, can be generalized to other aerosols in the wavelength range considered in
this work. Therefore, variations in the imaginary part of the refractive index have negligible influence on the WMLE technique.

However, as shown in Figure C.1, a small change in the real part of the refractive index causes obvious changes in the extinction coefficients. More specifically, an increase in the real part of refractive index causes the principal maxima of the extinction coefficients shift toward smaller $D_{32}$. This shift will cause a shift of the applicable range of a WMLE scheme toward smaller $D_{32}$ when applied to aerosols with larger real part of refractive index than that used in the design of the WMLE scheme, as illustrated in Figure 3.11. On the contrary, when applied to aerosols with smaller real part of refractive index than that used in the design, the applicable range of the WMLE scheme will be shifted toward larger $D_{32}$.

Next, the influence of refractive index on aerosol size measurement is investigated. Figure C.2 shows the calculation of the ratio of extinction coefficients between $\lambda_1=4 \mu m$ and $\lambda_2=0.5 \mu m$ for water aerosols following a log-normal distribution (with a distribution width of 1.4). According to Figure 3.9, this wavelength combination yields the optimized size measurement for $1.5 < D_{32} < 3.5 \mu m$. In Figure C.2, the ratio is first calculated using the refractive indices of water aerosol at a temperature of 22 °C at corresponding wavelengths, as shown in the caption of Figure C.2. Then the calculation is repeated with the real part of the refractive indices decreased and increased by 5%, respectively. As shown in Figure C.2, the variation in aerosol size measurement due to a 5% decrease in the real part of the refractive index is 0.61 μm (from 2.88 to 3.49 μm) at a ratio of 0.5. Therefore, quantitative understanding of the dependence of refractive index is required for accurate $D_{32}$ measurement. On the other hand, this calculation also indicates that if there is a method to independently measure and resolve such variation in aerosol diameter, then this independently measured variation can be used to infer the variation in the real part of the refractive index of the aerosols, and finally to infer the aerosol temperature after the relationship between refractive index of the aerosols and temperature has been quantified.
Finally, the dependence of the real part of the refractive on temperature needs to be considered. Refractive index of water has been studied extensively in a wide spectral range under different temperatures and pressures [1-3], and empirical formulations have been developed in certain wavelength, temperature, and pressure ranges [1]. These previous studies greatly facilitate the evaluation of temperature effects on the WMLE scheme. For example, Figure C.3 shows the variation of the real part of the refractive index of water from a temperature of 20 °C to 90 °C in a spectral range of 0.2 to 1.9 µm using the formula developed in [1]. The variation shown in Figure C.3 is defined by the following equation:

\[
\text{Variation} = \left| \frac{n_{90} - n_{20}}{n_{20}} \right| \times 100\%
\]  

(C.1)

where \(n_{20}\) and \(n_{90}\) are the real part of the refractive index at 20 and 90 °C, respectively. Figure C.3 suggests that variation in this spectral range is generally less than 1% except in the region near 0.2 µm. However, in other spectral regions water may exhibit stronger variation in the real part of refractive index. For example, at 3.413 µm, the real part of water’s refractive index varies by 1.5% when temperature changes from 16 to 50 °C, and varies 4% at a wavelength of 14.286 µm under the same temperature change [3]. In contrast to water, much less literature is available on the study of refractive index of other liquids [4, 5], therefore, variation of the refractive index with temperature can only be estimated at certain wavelengths. For example, the real part of isooctane’s refractive index varies by about 2.4% at a wavelength of 3.39 µm when temperature changes from 23 to 80 °C [5].

In applications where the aerosol temperature varies, such understanding of the dependence of the real part of the aerosol refractive index on temperature is highly valuable. If one desires to minimize the influence of temperature effect on aerosol measurements, wavelengths at which the real part of aerosol refractive is insensitive to temperature are preferred. On the other hand as discussed above, the wavelengths at which the real part of aerosol refractive exhibits enough sensitivity to temperature must be used for the
measurement of aerosol temperature.

References:


Figure C.1 Comparison of extinction coefficients at different refractive indices at a wavelength of 2 μm. Note that the difference between the calculation at $m=n-k\cdot i$ ($n=1.306$, and $k=0.0011$) and that at $m=n-1.25k\cdot i$ is too small to be distinguished and the curves corresponding to these calculations virtually overlap in this figure.

Figure C.2 Ratio of extinction coefficients at $\lambda_1=4$ μm and $\lambda_2=0.5$ μm for water droplets with a log-normal distribution ($\sigma=1.4$) at various refractive indices. Refractive index data: $n=1.335$ and $k=1\times10^{-9}$ at $\lambda=0.5$ μm and $n=1.351$ and $k=0.0046$ at $\lambda=4$ μm.
Figure C.3 Variation of the real part of the refractive index of water from 20 to 90 °C in the spectral range from 0.2 to 1.9 µm.
Appendix D
Calculation of Evaporation Rate Constant from Single Droplet Evaporation Model

The evaporation rate constant of isolated spherical droplets can be calculated from a theoretical model [1]. This appendix shows the calculation of the evaporation rate constant of water aerosols based on the model described in [1].

Evaporation rate constant of water droplet can be expressed by the following equation:

\[ k = \frac{8 \cdot k_G \cdot \ln(1 + B)}{c_{PG} \cdot \rho} \]  

(D.1)

where:

- \( k \) = evaporation rate constant
- \( k_G \) = thermal conductivity of the vapor mixture (background gas and water vapor)
- \( c_{PG} \) = specific heat at of the vapor mixture at constant pressure
- \( \rho \) = density of liquid water
- \( B \) = transfer number

Two transfer numbers, a mass transfer number (\( B_M \)) and a heat transfer number (\( B_T \)) as defined in Equations D.2 and D.3, can be used in Equation (D.1) to calculate \( k \).

\[ B_T = \frac{c_{PG} \cdot (T^\infty - T^S)}{c_L \cdot (T^S - T^\infty) + h_R} \]  

(D.2)

\[ B_M = \frac{Y_{H_2O}^S - Y_{H_2O}^\infty}{1 - Y_{H_2O}^\infty} \]  

(D.3)

where:

- \( T^\infty \) = ambient temperature
- \( T^S \) = surface temperature of the droplet
\[ T^0 \] = temperature of the droplet

\[ c_L \] = specific heat of liquid water

\[ h_{fg} \] = latent heat of vaporization of liquid water

\[ Y_{H_2O}^S \] = mass fraction of water vapor at the droplet surface

\[ Y_{H_2O}^\infty \] = mass fraction of water vapor in the background gas

\[ B_T \] and \[ B_M \] depend on droplet surface temperature (\( T_S \)). By invoking the assumption of unity Lewis number, \( B_T \) and \( B_M \) will be equal under steady-state evaporation, which allows the determination of droplet surface temperature. In this work, droplet surface is obtained by a graphical method, i.e., plotting graphs of \( B_T \) and \( B_M \) as a function of \( T_S \) finding the \( T_S \) at which these two curves intersect. After \( T_S \) is determined, it can be substituted into either Equation D.2 or D.3 to calculate the transfer number, which is then substituted into Equation D.1 to calculate the evaporation rate constant.

Here we provide an example calculation of the evaporation rate constant of water aerosols under the experimental conditions discussed in Section 5.4.2. In the experiment discussed in Section 5.4.2, the background gas is argon and the background temperature is 510 K. A surface temperature of 59 \(^\circ\)C and a transfer number of 0.0626 are determined, which leads to an evaporation rate constant of 0.019 mm\(^2\)/s. This calculated evaporation rate constant is in good agreement with the value inferred from measurement as discussed in Section 5.4.2.

References: