Combustion gas measurements using tunable laser absorption spectroscopy.

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Introduction

Increasing demands on combustion systems have stimulated new interest in combustion processes and in the development of laser-based diagnostics for combustion studies. This paper summarizes work at Stanford to develop and apply tunable infrared diode lasers for combustion gas measurements. These lasers are well suited for in situ measurements of species concentrations and temperature in combustion flows whenever a line-of-sight absorption measurement is appropriate. The same measurement techniques can be used to obtain fundamental spectroscopic data needed to describe the spectral characteristics of radiation from combustion systems such as engines and rockets.

The diode laser serves as a source of narrow-linewidth (< 10⁻⁴ cm⁻¹) infrared radiation whose wavelength can be rapidly modulated (> 10⁻³ cm⁻¹/ microsecond) to perform fast, high-resolution absorption spectroscopy. The complete fully-resolved absorption profile of a single vibration-rotation line can thus be quickly recorded, and from this one can infer the partial pressure of the absorbing species (stable species or radicals) and the lineshape parameters describing the absorption line. Temperature can be determined by measuring the relative absorption in adjacent lines originating from different vibrational levels. The ability to rapidly modulate the laser enables application of the same techniques to studies of non-steady combustion phenomena. Finally, the laser also is well suited for species and temperature measurements in particle-laden flows in that modulation of the laser wavelength on and off an absorption line allows simple discrimination against particle scattering and beam blockage effects.

Advantages of tunable diode laser absorption spectroscopy are its simplicity, high sensitivity, high spectral resolution (orders of magnitude improvement over conventional ir spectroscopy), and its fast modulation capability. The primary limitation, for some applications, is that it is a line-of-sight method. In recent work, we have demonstrated the feasibility of diode laser techniques for measuring species concentrations, temperature, and the spectroscopic parameters of line strength and collision halfwidths in the postflame region of a flat flame burner [1] and in a shock tube [2,3]. This paper includes examples from these previous flame studies and from new work involving laser absorption measurements in a fluctuating flame and in a particle-laden flame. The absorbing species in each of these cases is carbon monoxide, which was chosen for initial study because it is an important combustion species which has strong absorption and a simple ir spectrum. In the following sections, the experimental arrangement and relevant theory of infrared absorption are discussed, followed by a presentation of experimental results.

Experimental Arrangement

A schematic of the experimental arrangement is shown in Figure 1. The diode laser is a commercial system (Laser Analytics, Inc.) which includes a temperature-stabilized, closed-cycle refrigerator and a power supply to drive the laser. The laser elements, with final dimensions of approximately 0.2 x 0.3 x 0.5 mm, are fabricated from single crystal wafers of lead–salt compounds. Laser emission occurs from an area of about 10⁻⁴ cm². The nominal emission wavelength of each laser is determined by its composition, but the wavelength is also temperature dependent and hence may be tuned by varying the laser heat-sink temperature and/or bias current. The overall tuning range of individual lasers varies considerably; it is guaranteed to be greater than 50 cm⁻¹. Rapid modulation of the laser wavelength over narrow spectral intervals is performed by modulating the bias current.

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Figure 1. Optical arrangement for tunable diode laser spectroscopy in a flat-flame burner.

The output beam of the laser is collimated (or focused) to provide a spatial resolution of less than 3 mm as the beam passes through the flame 2-3 cm above the burner surface. After passing through a 1/2-meter grating monochromator (Jarrell-Ash, Ebert mounting), the laser beam is split into two beams, usually by means of an uncoated calcium fluoride window. The transmitted beam is sent
through a 10-cm absorption cell while the reflected beam is directed onto a solid germanium (2.5-μm length) Fabry-Perot etalon. Both beams are subsequently focused onto cooled (77 K), side-looking InSb detectors (photovoltaic, Judson J-10) with matching preamplifiers (Perry Model 720). Detector outputs are usually displayed simultaneously on a dual-trace oscilloscope.

The experimental procedure involves scanning the narrow-linewidth laser to record one or more fully-resolved vibration-rotation absorption lines in the molecule of interest, in the present case CO. Monochromator slits are typically set at 200 to 500 μm to obtain a single laser mode, although in some cases no slits are needed to achieve single-mode operation. In particular, the entrance slit is often removed since the laser image at the entrance slit is only about 250 μm. The laser power reaching detector D2 varies widely, depending on the laser element and the operating conditions. Single-mode power levels up to 100 μW have been observed in our experiments, but values of 10-20 μW are more typical. The monochromator is located on the detector side of the flame to filter radiation from the combustion gases. The absorption cell is used for wavelength calibration purposes and also to show the strong room-temperature absorption of transitions from the ground vibrational level of CO, that the laser is operating single mode. The cell is evacuated or removed while recording absorption lines in the flame.

The laser wavelength is modulated using a combined dc injection current and a variable amplitude, variable frequency sawtooth current, both provided as standard outputs of the Laser Analytics power supply. In the experiments reported here, with large fractional absorption (ΔI/I) and no significant noise problems, the absorption line profiles have usually been recorded on single-sweep oscilloscope traces with sweep rates ranging from 10-500 μsec/div. The detector-amplifier output is dc-coupled to the oscilloscope, with a variable dc-offset voltage E (see Figure 1) added in series so that the output voltage is zero (ground potential) when the laser beam is blocked. This arrangement eliminates the need for a separate recording of the zero-intensity level. When the fractional absorption is small, it is useful to display simultaneously both the ac-coupled and dc-coupled outputs on the oscilloscope. The gain on the ac-coupled channel can then be adjusted independently to compensate for variations in the fractional absorption. At very low absorptions, signal averaging of repetitive wavelength scans is useful to minimize the effects of noise. As an example, in a separate project involving measurement of NO in combustion flows, the detector-amplifier output is fed simultaneously to both an ac and a dc amplifier, the ratio of the amplifier outputs is formed using a divider circuit (to obtain ΔI/I), and the resulting absorption line profile is then sent to a signal averager. The result is a fully-resolved absorption line profile obtained under conditions where the peak absorption may be as small as 1%. A double-beam system is being developed to record even lower levels of absorption.

Two different burners have been used in the present experiments, a flat flame burner and a slot burner. Both systems are of conventional design and operate at atmospheric pressure with premixed fuel-air mixtures. The flat flame burner has a cross-section of 4 cm x 10 cm and is equipped with a ceramic-lined chimney and infrared-transmitting windows to ensure a constant optical path length through the absorbing gases. Fuel (methane or propane) and air flows are metered with calibrated rotometers and premixed before entering the base of the burner. A packed bed of glass beads in the burner base provides additional mixing and flow uniformity. The slot burner is a commercial unit (Perkin-Elmer atomic absorption burner, model 303) with slot dimensions of 0.5 mm x 5 cm. Rotometers are used to provide controlled fuel (C2H2) and air flows.

**Absorption Theory**

The theory required to interpret the experimental data is well established. The governing equation which links the measured transmissivity \( T_v \) to the absorbing species concentration and its absorption line parameters is the Bouguer-Lambert law of absorption

\[
T_v = \left( \frac{I}{I_0} \right)_v = \exp \left( -\int_0^L \beta_v P_{CO} \, dx \right)
\]

where \( I \) and \( I_0 \) are the transmitted laser intensities with and without absorption at wavenumber \( v \), \( P_{CO} \) is the partial pressure (atm) of the absorbing species, presently CO, and \( L \) is the pathlength (cm) across the flow. The absorption coefficient \( \beta_v \) is the product of the line strength \( S \) (cm⁻²-atm⁻¹) for the transition of interest and the lineshape factor \( \phi_v \) (cm); i.e., \( \beta_v = S \phi_v \),

where

\[
\int_0^L v \phi_v \, dv = 1
\]

Thus the line strength, sometimes called the line intensity, is simply the area under a curve of the absorption coefficient. In the present work, uniform conditions along the line of sight are assumed, so that the absorption law reduces simply to

\[
T_v = \exp (-\beta_v P_{CO} L)
\]

When absorption lines overlap, the relevant absorption coefficient is a summation: \( \beta_v = \sum S_l \phi_l \).

The line strength for a given transition in the fundamental band of CO (\( v' + 1 \to v'' + 1, J'' + 1 \)) is a known function of the temperature and the band strength evaluated at STP, \( S'(STP) \); i.e.,

\[
S_{v'v''+1} J''J + 1 = \left( S'(STP) / (273.2/T) \right) \sqrt{(v' + 1)} \end{align}
\]

\[
\exp[-(\Delta v_J J''hc/\kappa T)] \left[ 1 - \exp(-hvc/\kappa T) \right] S^J / Q(T)
\]

where \( S^J = J, P \) branch (\( J'' + J'' - 1 \))

\( S^J = J + 1, R \) branch (\( J'' + J'' + 1 \))

and \( Q(T) = v_J^J (2J + 1) \exp(-\Delta v_J J''hc/\kappa T) \)
is the partition function for vibration and rotation, \( T(v,J) \) is the energy of the \((v,J)\) state, in cm\(^{-1}\), \( v \) is the wavenumber for the specific transition, and \( \bar{v} \) is an average wavenumber, usually taken as the band-center value although small corrections can be calculated. The quantities \( h \), \( c \) and \( k \) are Planck's constant, the speed of light and Boltzmann's constant, respectively. For most purposes, it is sufficiently accurate (error of a few per cent or less) to use rigid-rotor, harmonic oscillator relations for the partition function. Recent determinations of the CO band strength have been in the range \( S^v = 260-280 \text{ cm}^{-2} \cdot \text{atm}^{-1} \) at STP (273.2 K).

The lineshape function \( \phi \), is defined assuming a Voigt profile which allows for a combination of Doppler and collision-line broadening. The Voigt function is tabulated in terms of the parameter \( a \), where

\[
a = (\ln 2)^{1/2} a \nu_D / \nu_D
\]

The Doppler-broadened linewidth (FWHM) is given by

\[
\Delta \nu_D = 7.16 \times 10^{-7} \left( \frac{T/M_{\text{CO}}}{1/\nu_0} \right)^{1/2} \nu_0
\]

where \( T \) is the temperature (K), \( M_{\text{CO}} \) is the molecular weight of the absorbing species (gm/mole), and \( \nu_0 \) is the line-center wavenumber. The collision-broadened linewidth is expressed in terms of the collision halfwidth \( \gamma \), i.e., the collision-broadened linewidth (FWHM) per unit pressure of the broadening species, and the pressure \( P \):

\[
\Delta \nu_C = 2\gamma (\text{cm}^{-1} \cdot \text{atm}^{-1})(P, \text{atm})
\]

The collision halfwidth for a given transition is a function of temperature and the broadening species. In the present diode laser experiments, the temperature (and hence \( \Delta \nu_c \)) is usually known so that it is straightforward to infer values for the parameters \( a \) and \( \Delta \nu_c \), and hence \( \nu_D \) and \( 2\gamma \), from the observed absorption linewidth.

In the past, most determinations of collision halfwidth have been made near room temperature, and high-temperature values have been obtained by extrapolation, usually assuming a \( T^0.5 \) temperature dependence so that

\[
2\gamma = 2\gamma^* (300/T)^{0.5}
\]

where \( 2\gamma^* \) is the collision halfwidth at 300 K. This temperature dependence is based on hard-sphere collision theory arguments and is almost certainly in error, though to an unknown degree. The current lack of high-temperature data for collision halfwidths, together with the accuracy and simplicity of diode laser measurements of linewidths in flames and shock-heated flows suggests that this may become a fruitful area of diode laser research.

The above relations for line strength and lineshape have been combined to provide plots of CO line-center absorption coefficients versus temperature in Figure 2. Results are shown for a limited number of P-branch transitions with \( v'' = 0 \) and \( v'' = 1 \). The calculations are for a pressure of 1.0 atm and assume purely collision-broadened lines.

This plot enables a useful estimate of the detection limits of laser absorption for measuring CO concentrations in atmospheric-pressure, high-temperature gases. As an example, the absorption coefficient for the \( v'' = 0 \), \( P(20) \) line of CO at \( T = 2000 \text{ K} \) is \( \beta = 5 \text{ cm}^{-1} \cdot \text{atm}^{-1} \). Assuming a path length of 10 cm, and a minimum detection limit of 0.1\% absorption, the minimum detectable mole fraction of CO is calculated to be \( 2 \times 10^{-5} \) (20 ppm). This limit should be adequate for most combustion studies. In cases where the CO concentration is large or the path length is long, it may be desirable to use a transition with a smaller absorption coefficient to avoid saturation at line center. The strong temperature dependence of some \( v'' = 0 \) transitions can also be a problem, particularly in measurements on flows with a hot core and cold boundary layers. Increased absorption in such layers can be avoided, if necessary, through proper choice of the absorption line. Lines originating from excited vibrational levels, \( v'' = 1 \) and higher, are attractive in this regard.

![Figure 2. Calculated line-center absorption coefficients for CO vs. temperature at a pressure of 1.0 atm. A collision-broadened lineshape is assumed with \( 2\gamma^* = 0.1 \text{ cm}^{-1} \cdot \text{atm}^{-1} \). The band strength assumed is \( S^v (273.2) = 260 \text{ cm}^{-2} \cdot \text{atm}^{-1} \).](image)

Flat Flame Results

Measurements of CO species concentrations, collision halfwidths and temperature have been made in the postflame region of a flat flame burner. Some of this work has been reported previously. This facility provides a uniform, well-characterized combustion flow which is nearly ideal for initial experiments aimed at validating the laser absorption technique by comparison with probe-based measurements (sampling probe and thermocouple) and theoretical predictions based on metered flow rates.

A typical data trace used to infer both the collision halfwidth and the CO concentration is
shown in Figure 3. This is a single-sweep oscillogram, recorded at 200 μsec/div, of the ν = 2 → 1, P(10) transition at ν = 2077.0 cm⁻¹. The upper trace records the power transmitted by the Fabry-Perot etalon (0.0495 cm⁻¹ free spectral range) and provides a direct measurement of the change in laser frequency. The lower traces record the transmissivity as the laser is tuned across the absorption line. The zero-intensity trace is recorded by resewing the scope after either blocking the laser beam or grounding the scope input.

Figure 3. Oscillogram of fully resolved CO absorption line at ν = 2077.0 cm⁻¹ (ν = 2→1, P(10)) in an atmospheric pressure propane-air flat flame burner.

The fuel-air equivalence ratio of this fuel-rich flame is Φ = 1.37, and the adiabatic flame temperature is 2068 K. The temperature at the optical axis (2 cm above the burner surface) is 1974 K, measured using a Pt/Pt - 13% Rh thermocouple (0.13-mm diam. wire) corrected for radiation loss. The calculated partial pressure of CO, based on the metered flow rates and assuming chemical equilibrium at the measured temperature, is 0.084 atm. At these CO concentrations, line-center absorption is nearly complete for accessible transitions from ν' = 0, so that transitions from ν' = 1 have been used.

The absorption lineshape for a similar condition was previously shown to be consistent with a Voigt profile. In the present case, a good fit to the absorption lineshape is obtained using a value of 2.50 for the Voigt parameter a. This value of a is determined from the measured linewidth (FWHM = 0.0416 cm⁻¹) and the calculated Doppler linewidth (ΔνD = 0.0125 cm⁻¹) using Voigt function tables. The collision linewidth, calculated from ΔνC and a, is ΔνC = 0.0376 cm⁻¹. Since the experiment is performed at a pressure of 1.0 atm, this latter figure is also the collision halfwidth for the absorption line under these combustion conditions, i.e., 2γ(1974 K) = 0.0376 cm⁻¹ - atm⁻¹. The corresponding room temperature collision halfwidth, calculated assuming a T⁻⁰.5 temperature dependence, is 0.096 cm⁻¹ - atm⁻¹. This result is very close to that reported earlier for the same transition.

Further experiments to determine the dependence of the collision halfwidth on temperature and also on the vibrational and rotational quantum numbers are planned.

Once the value of a is determined from an absorption record, it is straightforward to convert a measurement of the transmissivity, for example at line center, to a value for the partial pressure of CO. For the absorption record in Figure 3, the transmissivity at line center is Tν = 0.262 and the lineshape function f(0) = 16.0 cm⁻¹, the latter quantity being obtained from tabulations of the Voigt function where a = 2.50. The remaining quantities required to solve the Bouguer-Lambert equation for TCO are the path length l = 10.0 cm and the line strength S(1974 K) = 0.107 cm⁻¹ (based on a band strength S(Δν) = 260 cm⁻² - atm⁻¹). The resulting value for TCO is 0.079 atm which agrees within experimental error with the value calculated from the metered fuel and air flows and the temperature.

The flat flame burner also provides a useful test bed for developing a temperature measurement technique based on tunable laser absorption spectroscopy. In brief, the proposed technique involves tuning the laser wavelength to measure the relative absorption in two nearly coincident vibration-rotation lines originating from two different vibrational levels of the same ir-active species. Details of the technique and the required theory have been described previously.

A sample temperature measurement is shown in Figure 4. The format of the oscillogram is similar to that for species or halfwidth measurements, except that two absorption lines are displayed, in this case the ν = 3→2, P(3) and ν = 2→1, P(3) lines near 2105.2 cm⁻¹. The line separation is 0.131 cm⁻¹. This particular single-sweep oscillogram was recorded at 200 μsec/div (a laser modulation rate of 500 Hz), but faster recording up to 10 μsec/div (laser modulation rate of 10 kHz) has been demonstrated for the same line pair. Where appropriate, the repetitive signals can be time averaged for improved accuracy. The temperature inferred from the ratio of line-center absorptions displayed in Figure 4 is 2100 K, while the temperature measured using a radiation-corrected thermocouple is 2070 K. The difference in temperature is within the experimental error of the technique.

Since the predominant combustion species is N₂, the value of 2γ inferred here can be compared with that obtained in room-temperature absorption cell experiments conducted with CO in N₂ in which [f(ν = 0, P(10))]₂γ was found to be 0.116 cm⁻¹ - atm⁻¹.11 This difference in 2γ may be attributed to a combination of factors, including possible decreased broadening by other combustion species, dependence of 2γ on ν', or as is most likely, the use of an incorrect temperature dependence for 2γ. If the difference is assumed due to temperature dependence alone, then the room-temperature and high-temperature values for 2γ can be used to determine a new temperature dependence, with the result

\[ 2γ = 2γ'(300/K)^{0.60} \]

Further experiments to determine the dependence of the collision halfwidth on temperature and also on the vibrational and rotational quantum numbers are planned.
uncertainty of each measurement (approximately ± 40 K).

\[ N_{v,J} = \frac{(2J+1)/Q(T)}{\exp[-T(v,J)/h\nu kT]}. \]

where \( T(v,J) \) is the energy of the state and \( Q(T) \) is the partition function for vibration and rotation.

Plots of \( S_1/S_2 \) vs. temperature for a number of line pairs in CO have been reported previously.\(^2\)

The sensitivity of the temperature measurement is determined by the slopes of these curves. As an example, for the line pair employed in Fig. 4 and a gas temperature of 2000 K, a 1% change in \( S_1/S_2 \) (and hence also in the measured quantity \( R \)) corresponds to a temperature change of only 0.64% (13 K).

This temperature measurement technique is not limited to a few fortuitous coincidences of lines. A number of favorable line pairs exist for CO, and in addition the same approach can be used with other combustion species such as CO\(_2\). The optimum line pair for determining temperature will of course depend on the experimental conditions, the species present and the availability of diode lasers. In general, one would choose lines with easily measured transmissivities (e.g., 0.1 < \( T_v < 0.9 \)) to enable an accurate determination of \( R \), and with a large slope for \( S_1/S_2 \) vs. \( T \) to reduce the uncertainty in \( T \) associated with the experimental uncertainty in \( R \). Small values of \( S_1/S_2 \) imply potential difficulty in measuring the transmissivities since the values for \( T_v \) may be near 0 or 1. The separation between lines may also be important. The lines should be sufficiently close to fall within the tuning range of a single laser mode and yet not so close as to have excessive overlap. Some overlap can be easily accounted for in the data analysis, however, by calculating the lineshape function for the observed linewidths and line separation.

### Slot Burner Results

New diode laser results obtained using a slot burner are reported in this section. The objectives of these experiments were to demonstrate laser absorption measurements under fluctuating combustion conditions and in particle-laden flows.

In the first set of experiments, a rotating valve was installed in the air line feeding the mixing section of the slot burner. This valve operated at 60 Hz, producing repeatable fluctuations in the equivalence ratio with a period of 16.7 msec.

Typical results in a fuel rich flame are shown in Fig. 4. The top panel, Fig. 5a, displays several sweeps of an individual CO line \((v'' = 0, P(7))\) at \( \nu = 2115.6 \text{ cm}^{-1} \) taken at different times in the fluctuation cycle. The sweep time for each trace is about 1 msec, which is sufficiently short that each absorption line trace may essentially be viewed as a snapshot or instantaneous record. The middle panel, Fig. 5b, was recorded with a slow oscilloscope sweep (10 msec/div.) so that the absorption line was scanned 28 times in each fluctuation cycle. (Note that one modulation cycle of the laser involves two sweeps across the line, once as the current is ramped up and once as the current is ramped back to its initial value.) The oscillogram in Fig. 5b is thus a compressed recording of approximately 150 absorption line scans. Fig. 5c is simply a recording of the time-varying absorption with
in Fig. 5a. We conclude that the fluctuations in absorption, for example at line center, are primarily due to variations in the CO concentration in the observation region.

The laser absorption technique can be applied to flows with a wide range of time scales. The laser modulation rate can be varied simply by charging the repetition rate of the current waveform. Modulation rates of greater than 10 kHz have been demonstrated\textsuperscript{2,4} for a laser tuning range of greater than 0.5 cm\textsuperscript{-1}. In many cases, oscilloscope recording will be insufficient and a system providing both time resolution and long-duration recording capability may be needed.

The slot burner also has been used to perform laser absorption experiments in sooting combustion flows. The objective in this work is to demonstrate the utility of tunable laser absorption as a measurement technique in dirty or particle-laden combustion flows. Operation of the slot burner on rich C\textsubscript{2}H\textsubscript{2}-air mixtures produces a convenient means of generating realistic submicron-sized particles (soot). Recent experiments under similar operating conditions\textsuperscript{12} suggest that the particle diameter is in the range 0.1-0.3 microns. Under these conditions, the combustion gases appear opaque, even in the short dimension (1 cm) of the flame transverse to the optical axis of the laser measurement. The flame radiation varied from intense yellow to yellow-white depending on the fuel-air ratio.

A typical laser absorption record obtained in a sooting flame is shown in Fig. 6. The format of the record is similar to that shown previously, except that a baseline trace without absorption or scattering (labelled "no flame" in Fig. 6) is also shown. The difference between the "no flame" and "flame on" traces represents the attenuation of the laser beam due to scattering by soot particles. If the effective scattering area of the particles is known, the fractional attenuation can be used, together with the path length (5 cm), to calculate the number density of soot particles. Using the geometrical area of 0.25 micron diam. particles as a probable upper bound for the scattering area, the lower bound on the number density is found to be 10\textsuperscript{8} particles/cm\textsuperscript{3}.

The objective of these experiments was not to analyze this particular fluctuating combustion flow in detail, but rather to demonstrate the variety of information which a tunable laser can provide in studying nonsteady flows. A series of laser temperature measurements was also recorded for this flow, however the temperature fluctuations observed were relatively small. This is also consistent with the small variations in absorption linewidth displayed.
The significance of these measurements is that they confirm the usefulness of tunable laser absorption techniques for measuring species concentrations and temperature in particle-laden combustion flows. As long as sufficient laser radiation is transmitted through the flow, the use of wavelength modulation provides a simple means of discriminating against the continuum attenuation due to particles. The absorption records show no perceptible increase in noise and can be reduced in exactly the same manner as those for clean flows.

The data in Fig. 6 were obtained using the \( \nu'' = 3, R(21) \) transition of CO at \( \nu = 2137.20 \) cm\(^{-1}\). The observed linewidth is 0.041 cm\(^{-1}\), which is nearly the same as found previously in the flat flame experiments. Unfortunately, the temperature was not measured directly, but the linewidth suggests a temperature of about 2000 K, which seems reasonable for this rich acetylene flame. The partial pressure of CO is calculated to be 0.11 atm.

**Concluding Remarks**

These experiments demonstrate that tunable diode lasers are well suited for in situ measurements of species concentrations and temperature in combustion flows whenever a line-of-sight absorption measurement is appropriate. The techniques also are applicable to nonsteady and particle-laden flows.

Tunable diode laser measurements of fully resolved absorption lines in flames provide an important new source of high-temperature spectroscopic data for collision halfwidths and line strengths. Such data should prove useful both in characterizing the radiation from practical devices (such as engines and rockets) and in testing theories used to predict these quantities at high temperatures.

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**References**


8. See Ref. 7, Chapter 7, Section 5, and the references listed there.


