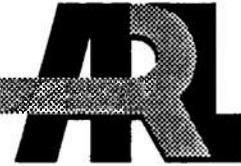




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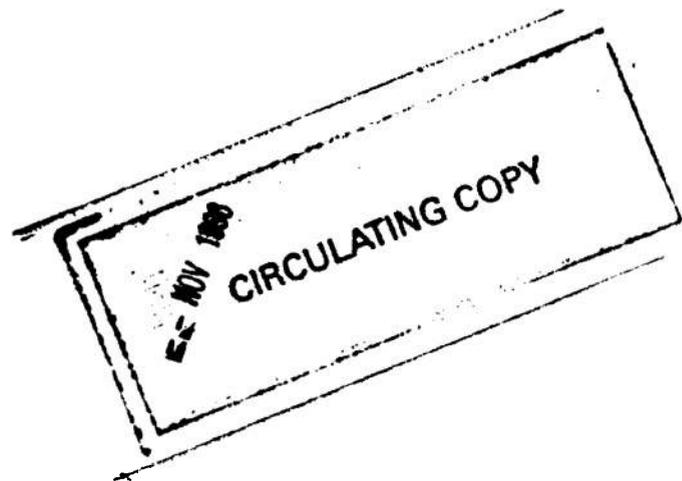
A Novel Flame Thermometer Based on the Doppler Width of Ro-Vibrational Transitions

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13. ABSTRACT (Maximum 200 words) A novel flame thermometric technique based on the infrared spectroscopy of flame species is reported. It involves the use of a narrow linewidth ($<0.001 \text{ cm}^{-1}$) tunable infrared diode laser to measure fully rotationally resolved absorption spectral profiles associated with the following carbon monoxide (CO) transitions: 1) $v = 1 \leftarrow 0$ P23 at $2,046.2765 \text{ cm}^{-1}$, 2) $v = 2 \leftarrow 1$ P19 at $2,038.5820 \text{ cm}^{-1}$ and 3) $v = 2 \leftarrow 1$ P21 at $2,029.6560 \text{ cm}^{-1}$. The CO line profiles are fitted to a Voigt function using a multi-parameter least-squares fitting routine and the temperature is calculated from the half-width of the Doppler component of the Voigt linewidth. Temperature profiles of premixed $\text{C}_2\text{H}_4/\text{O}_2/\text{Ar}$ and $\text{CH}_4/\text{O}_2/\text{Ar}$ flames were determined from the Doppler width of CO transitions recorded at various heights above the burner surface. Comparisons are made to temperature profiles obtained with $190\text{-}\mu\text{m}$ -diameter coated Pt/Pt-Rh(10%) thermocouples.			
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1. INTRODUCTION

A complete understanding of the complex network of chemical reactions occurring in flame environments requires the determination of spatially resolved species concentration and temperature profiles which, in turn, can be used as stringent tests for present and developing flame models. Temperature profiles have traditionally been measured using fine wire thermocouples, which can provide a high degree of spatial resolution in a flame given that thermocouple diameters are on the order of ten to hundreds of microns. The thermocouple, however, constitutes a physical interference which may alter the local flame environment. To minimize perturbations to the flame, thermocouples are made as small in diameter as possible and are often coated to reduce or eliminate surface catalytic effects. Another advantage of keeping the diameter of the thermocouple as small as possible is that corrections made to the measured temperature for radiation losses and heat conduction are minimal.

Recently, a number of non-intrusive spectroscopic techniques have been developed for combustion diagnostics. In particular, laser-based diagnostic techniques have the high sensitivity, spatial resolution, and species selectivity needed to measure spatially resolved species concentration and temperature profiles in flames. Considerable efforts have gone into using laser-induced fluorescence (LIF) of the hydroxyl radical as a flame thermometer (Rensberger et al. 1989) because the spectroscopy of the diatomic OH radical is well understood and OH is ubiquitous in flame environments. For example, LIF measurements of the hydroxyl radical via the $A^2\Sigma^+ \leftarrow X^2\Pi$ (1,0) band near 281 nm have been used to determine temperatures in a low-pressure, premixed $C_2H_4/O_2/Ar$ flame (Sausa et al. 1990).

Spectroscopic techniques, however, often involve the use of highly energetic ultraviolet (UV) photons delivered from lasers at a high fluence. The absorption of the probe radiation by other flame species can result in photodissociation which can lead to errors in the measured local flame temperatures and species concentrations. This is especially significant in propellant-like flames in which NO_2 is the primary oxidizer since NO_2 is photolyzed at wavelengths less than 405 nm (Calvert and Pitts 1967). Consequently, a diagnostic tool which does not alter the local flame environment is sought to monitor the combustion processes in low-pressure flames. One such technique is infrared spectroscopy.

Tunable Pb:salt diode lasers are an excellent source of narrow linewidth ($<0.001\text{ cm}^{-1}$) infrared radiation which can be rapidly scanned to perform fast, high-resolution absorption spectroscopy. With the advantages of high sensitivity and high resolution, tunable diode laser spectroscopy is a potentially

powerful diagnostic technique whenever a line-of-sight method is suitable. In the late 1970s, Hanson and co-workers introduced infrared diode laser spectroscopy as a combustion diagnostic technique. In these studies, the absorption of infrared radiation by CO and NO in a gas cell, shock tube, and various flame environments was investigated. These absorption measurements yielded quantitative CO (Schoenung and Hanson 1981; Hanson 1983) and NO (Hanson 1983; Falcone, Hanson, and Kruger 1983) concentrations and temperatures (Hanson and Falcone 1978) were determined from the relative intensity of two closely spaced ro-vibrational transitions originating from different vibrational levels.

This report describes the novel use of a tunable infrared diode laser to measure the temperature profiles of low pressure (20 torr) premixed flames. Rather than using the two-line method described in the literature (Hanson and Falcone 1978), the local flame temperature is determined from the Doppler width of a single CO absorption spectral profile. To verify this method as a suitable flame diagnostic technique, the temperature profiles of premixed $C_2H_4/O_2/Ar$ and $CH_4/O_2/Ar$ flames were derived by recording fully resolved CO ro-vibrational transitions at various heights above the burner surface. The measured CO absorption spectral profiles are fitted to a Voigt function using a multi-parameter least squares fitting routine and the temperature is calculated from the Doppler component of the Voigt linewidth. Comparisons are made to temperature profiles obtained with a 190- μm -diameter Pt/Pt-Rh(10%) thermocouple coated with beryllium oxide (15%)/yttrium oxide which eliminates surface catalytic effects.

2. EXPERIMENTAL

The low-pressure burner chamber constructed at the U.S. Army Research Laboratory (ARL) is equipped with a number of combustion diagnostic techniques for investigating one-dimensional premixed laminar flow flames. These include molecular beam sampling with triple quadrupole mass spectrometric detection, LIF, resonance enhanced multiphoton ionization, and temperature measurements by thermocouple and laser-based techniques. The experimental apparatus has been described in detail elsewhere (Sausa et al. 1992), however, a brief description of the experiment pertinent to the present study is given below. Figure 1 shows a schematic representation of the low-pressure burner chamber and tunable diode laser system. The low-pressure burner chamber is equipped with a flat flame McKenna burner mounted on a motorized high-vacuum feed through which facilitates vertical scanning of the burner. The chamber pressure is measured and regulated with a MKS pressure transducer interfaced with a MKS pressure controller and exhaust throttle valve. The flames investigated in this study were stabilized at operating pressures of 20 torr with gas flow rates regulated with calibrated Tylan and MKS mass flow

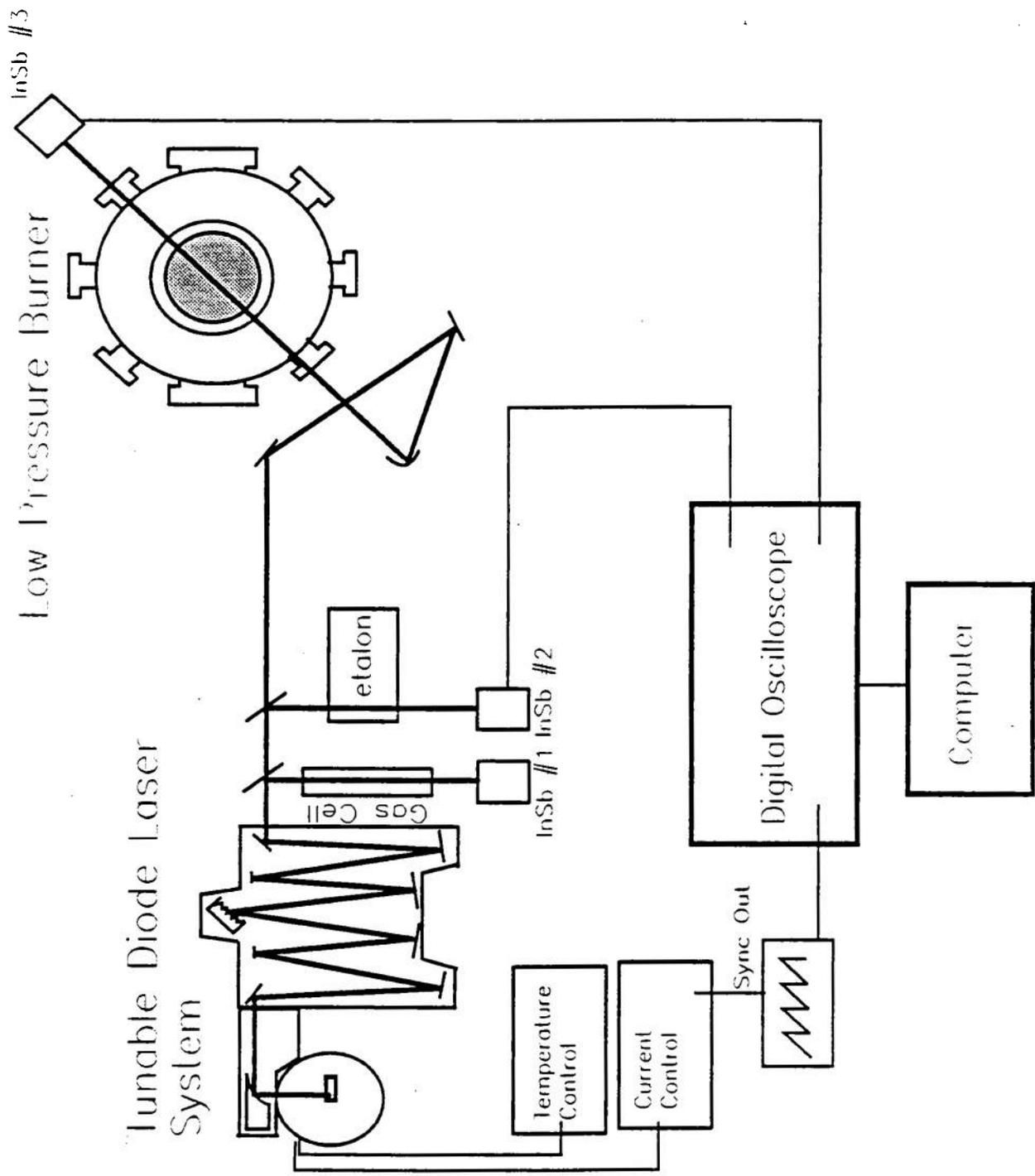


Figure 1. A schematic representation of the tunable diode laser system and low-pressure burner chamber.

controllers, ranging from 0.40 to 2.80 standard liters per minute. Identical flame conditions were established for corresponding thermocouple and Doppler temperature measurements. The gases were standard high-purity grade and used without further purification.

The tunable diode laser system (Laser Photonics, Inc. - Analytics Division) currently contains three laser diodes. Each laser diode is a small, single crystal lead salt chip seated in a gold-plated copper package designed for mounting to a cold finger in a liquid nitrogen dewar. The lowest achievable temperature is approximately 80 K, and the high temperature operating limit depends on the particular laser diode. Tunable infrared radiation is available in the following frequency ranges from the three currently installed laser diodes: 1) $2,055.2 \pm 50 \text{ cm}^{-1}$ for exciting the CO stretching vibration; 2) $1,921.1 \pm 25 \text{ cm}^{-1}$ for exciting the NO stretching vibration; and 3) $2,889.0 \pm 10 \text{ cm}^{-1}$ for exciting the $\text{NO}_2 \nu_1 + \nu_3$ combination band. The CO laser diode was used to obtain the experimental results presented in this study.

Output radiation from the laser diode is incident onto a parabolic aluminum mirror which collimates the beam and directs it into a monochromator compartment and onto a diffraction grating for frequency discrimination. The tuned infrared radiation is then directed through two pellicle beamsplitters which reflect approximately 10% of the main beam through a 15-cm-long static gas cell and a passive confocal etalon, respectively. The confocal etalon consists of two gold-coated spherical mirrors spaced 25 cm apart such that the free spectral range of the cavity is roughly 0.01 cm^{-1} . Both reflected beams are detected with separate liquid nitrogen-cooled InSb detectors. The transmission spectrum of the sample in the gas cell provides an absolute reference frequency and the etalon fringes provide a relative frequency calibration. The main beam is directed through the low-pressure burner chamber, parallel to the burner surface, and onto a third liquid nitrogen-cooled InSb detector. The diameter of the focused beam in the chamber is currently $\sim 2 \text{ mm}$ with a laser linewidth on the order of 10–20 MHz (FWHM).

To record a given spectral profile, the laser current is modulated with a positive saw tooth waveform over a short range (typically 10 mA which corresponds to 0.1 cm^{-1}) at high frequency (200 Hz) and fixed laser diode temperature. The initial current is adjusted such that the frequency of the CO transition of interest is located in the center of the sawtooth ramp. The resulting spectral and etalon fringe traces are monitored on a digital oscilloscope (LeCroy 7200), averaged for 100 scans, and transmitted over a GPIB interface to a microcomputer for data processing and analysis. In this way, a fully calibrated spectral scan of up to 0.5 cm^{-1} can be recorded in less than 1 minute.

The thermocouples used in this study were constructed from 125- μm -diameter platinum and platinum with 10% rhodium wires spot-welded together to produce a thermocouple junction. The thermocouple wire was mounted on a V-shaped holder and a spring was used to keep the wire taut, which eliminated any sagging of the wire in the high-temperature flame environment. The thermocouple wire was coated with a beryllium oxide (15%)/yttrium oxide mixture following the procedure described in the literature by J. H. Kent (1970) to avoid surface catalytic effects. Temperature was measured by scanning the burner in both the forward and reverse directions relative to the fixed thermocouple junction. The average temperature was then corrected for radiation losses from the 190- μm -diameter coated thermocouple junction (Sausa et al. 1992). The absolute temperature measurements have an uncertainty of ± 50 K in the burnt gas region of the flame where the temperature is high and the radiation correction is large (200 K). The uncertainty in the cooler preheat region of the flame is ± 20 K.

3. THE SPECTRAL LINESHAPE MODEL

The measured CO line profiles were fitted to a Voigt profile using a multi-parameter least-squares fitting routine. The Voigt profile is a convolution of a Gaussian and a Lorentzian function, corresponding to the inhomogeneous (Doppler) and homogeneous (collisional) contributions to the line profile, respectively. The laser linewidth is ignored in the model since it is an order of magnitude smaller than the other contributions to the linewidth. The Voigt function is defined in the following expression in terms of the Voigt parameter, a , which is proportional to the ratio of the collisional and Doppler widths, and the normalized distance from the line center frequency.

$$F(\nu - \nu_0) = (2/\Delta\nu_D)(\ln 2/\pi)^{1/2}V(a, x)$$

where,

$$V(a, x) = \frac{a}{\pi} \int_{-\infty}^{\infty} \frac{e^{-y^2} dy}{a^2 + (x - y)^2}$$

$$a = (\ln 2)^{1/2}(\gamma_L/\Delta\nu_D) \quad \text{and} \quad x = (\ln 2)^{1/2}(2/\Delta\nu_D)(\nu - \nu_0) .$$

The integral of the Voigt function over frequency space must be solved numerically since it does not have an analytical solution. Temperatures are determined from the Doppler component of the Voigt linewidth according to the following expression (Mitchell and Zemansky 1971):

$$\Delta\nu_D = 7.162 \times 10^{-7} (T/m_{\text{CO}})^{1/2} \nu_0$$

where $\Delta\nu_D$ is the Doppler width (FWHM) in wavenumbers, T is the temperature in Kelvin, m_{CO} is the molecular weight of the absorbing species (in this case, CO), and ν_0 is the center frequency of the measured transition in wavenumbers.

The temperature dependence of the collisional broadening parameter (γ_L) is also included in the model using the following well-known relationship derived from the Anderson-Tsao-Curmutte (ATC) theory of spectral lineshapes (Tsao and Curmutte 1962):

$$\gamma_L(T) = \gamma_L(T_{\text{ref}}) (T/T_{\text{ref}})^{-N}$$

By including this relationship in the model, several assumptions are made concerning the input parameters $\gamma_L(T_{\text{ref}})$, T_{ref} , and N as described below.

The collisional broadening parameter, $\gamma_L(T)$, is very sensitive to the rotational (J) quantum number of the measured transition, as well as to the collision gases present in the sample. Collisional broadening parameters for CO have been measured for a number of J states in the fundamental ($\nu = 1 \leftarrow 0$) and first overtone ($\nu = 2 \leftarrow 0$) bands over a limited temperature range (Varghese and Hanson 1980; Soufiani and Hartman 1987; Bouanich and Haeusler 1972; Nakazawa and Tanaka 1981; Lowry and Fisher 1982). Two of the CO transitions of interest to the present study are hot band ($\nu = 2 \leftarrow 1$) transitions. These transitions were chosen because there is significant population in the first vibrational level at the higher temperatures exhibited in the flame. Therefore, by monitoring hot band transitions, the errors caused by edge effects, which can limit a line-of-sight technique, can be limited. The collisional broadening parameters of these hot band transitions, however, have not been accurately determined. While the collisional broadening parameter is sensitive to the rotational state of the absorbing species, it is relatively insensitive to the vibrational state. In this study, the vibrational state dependence of $\gamma_L(T)$ is assumed to be negligible (Varghese and Hanson 1980) and the collisional broadening parameters for CO in the appropriate J states in the ground vibrational state were used in the fits to the data.

As stated above, the collisional broadening parameter for CO is sensitive to the collision gases present in the sample. For a mixture of gases, the collisional broadening parameter for the absorbing species of interest is equivalent to the sum of the products of the collisional broadening parameters for each

individual collision partner and their mole fractions in the mixture. In a flame environment, there are numerous possible collision partners, from reactants to radical intermediates to products, and the composition of the gases in the flame is constantly changing as a function of height above the burner. Without some a priori determination of the actual gas composition at each point in the flame, it is difficult to assign a precise value to $\gamma_L(T_{ref})$. As a first approximation, however, the collisional broadening parameters for CO-Ar interactions are chosen as input parameters in the model since the flow rate of Argon (2.8 slm) constituted more than 50% of the premixed gases present in the C₂H₄/O₂/Ar and CH₄/O₂/Ar flames investigated.

There has been considerable discussion in the literature about the correct value of N to use in the above expression for the temperature dependence of the collisional broadening (Varghese and Hanson 1980; Soufiani and Hartman 1987; Bouanich and Haeusler 1972; Nakazawa and Tanaka 1981; Lowry and Fisher 1982). The temperature exponent, N, typically ranges from 0.50 (in the hard sphere limit) to 1.0 and varies with the temperature and rotational state of the absorbing species. Over the narrow range of J values for the CO transitions measured in the present study, N does not change significantly (Bonamy, Robert, and Boulet 1984) and is input as a constant value of 0.70. The assignments and transition frequencies for the three CO transitions measured in the C₂H₄/O₂/Ar and CH₄/O₂/Ar flames, as well as the values of $\gamma_L(T_{ref})$, T_{ref} and N used as input parameters in the fits to the data, are listed in Table 1. These input parameters were taken from the literature and were not varied in the fits to the data.

Table 1. Input Parameters for the Model Used in the Fits to the Absorption Profiles of CO Measured in Low-Pressure Flames

CO Transition	Center Frequency (cm ⁻¹)	$\gamma_L(T_{ref})$ (cm ⁻¹ /atm)	T _{ref} (K)	N
v = 1←0 P23	2,046.2765	0.079 ^a	300	0.70 ^b
v = 2←1 P19	2,038.5820	0.079 ^a	300	0.70 ^b
v = 2←1 P21	2,029.6560	0.071 ^a	300	0.70 ^b

^a Varghese and Hanson (1980).

^b Bonamy, Robert, and Boulet (1984).

4. RESULTS AND DISCUSSION

Shown in Figure 2 is the CO spectral profile associated with the $v = 1 \leftarrow 0$, P23 transition at $2,046.2765 \text{ cm}^{-1}$ recorded in a 20-torr $\text{C}_2\text{H}_4/\text{O}_2/\text{Ar}$ flame 8.2 mm above the burner surface, which is ~ 3 mm above the flame front in the post-flame region. The data points represent the experimental spectral profile while the solid line is the result of fitting the data to the Voigt profile described above. The resulting temperature determined from the fit is $1,799 \pm 3 \text{ K}$, where the quoted uncertainty is one standard deviation of the fitted value. The uncertainty in the fitted temperature, however, is also dependent on the input collisional broadening parameter, which is not well defined for the wide range of temperatures and collision gases present in the flame. Given the above assumptions, a 10% change in N or a 10% change in $\gamma_L(T_{ref})$ changes the fitted temperature by approximately 2 K and 18 K, respectively. The accuracy of the method is estimated to be approximately 50 K resulting from the uncertainties placed on the input model parameters based on the stated assumptions.

The temperature profile of a 20-torr $\text{C}_2\text{H}_4/\text{O}_2/\text{Ar}$ (0.4/1.2/2.8 slm) flame derived from the Doppler width of the three CO transitions listed in Table 1 are compared to Pt/Pt-Rh(10%) thermocouple measurements in Figure 3. The temperatures determined from the Doppler width of the two hot band transitions agree well with the thermocouple measurements at positions in the flame high above the burner surface. The fact that the two hot band transitions do not yield the same temperature at a given height above the burner may be an indication that the collisional broadening parameters for the two transitions used as input in the fits to the data are in error. However, as can be seen in Figure 3, the temperatures determined from the two hot band transitions are similar within the error limits placed on the fitted temperatures due to the assumptions made in the model function ($\pm 50 \text{ K}$).

The temperatures determined from the Doppler width of the $v = 1 \leftarrow 0$ transition are consistently much lower than the thermocouple results. The collisional broadening parameter would have to be in error by more than 100% to cause such an error in the fitted temperature. We consider this unlikely. The error in the temperatures determined from the Doppler width of the fundamental band transition is most likely caused by interference of "cold" CO that is present beyond the edges of the burner surface. This problem has been minimized by inserting windows into the chamber approximately 5 mm away from the edges of the burner head and by flowing Ar gas up from the bottom of the chamber to flush away any recirculated gases. However, the problem has not been eliminated. Also evident in Figure 3 is that all three temperature profiles determined from the CO Doppler width peak 5 mm closer to the burner head

CO Line Profile

$\nu=1-0$ P23 $2046.2765 \text{ cm}^{-1}$

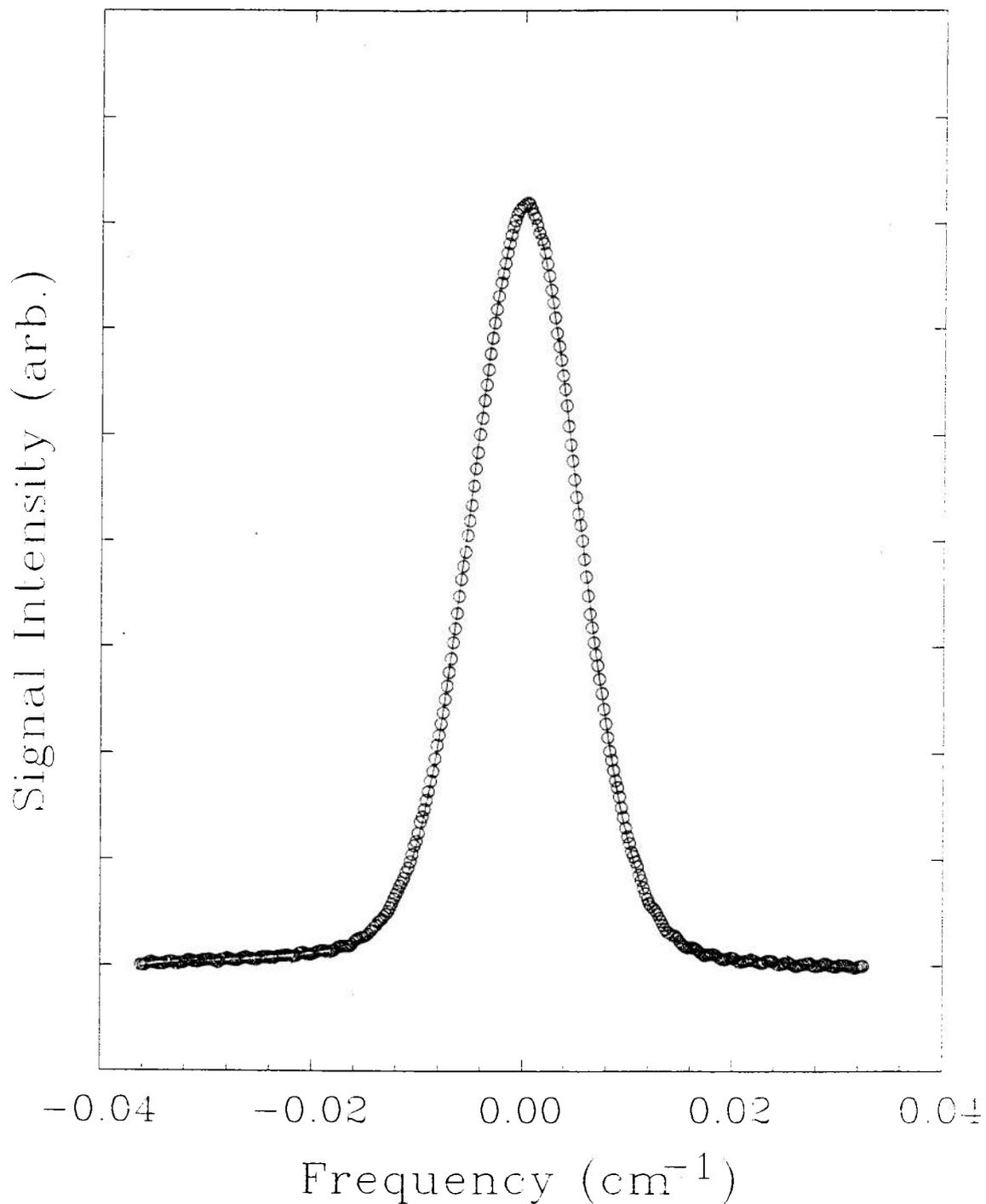


Figure 2. Observed and fitted CO absorption profile measured 8.2 mm above the burner surface in a 20-torr stoichiometric $\text{C}_2\text{H}_4/\text{O}_2/\text{Ar}$ (0.4/1.2/2.8 slm) flame. The fitted temperature is $1,799 \pm 50$ K.

$C_2H_4/O_2/Ar$ Flame

0.4/1.2/2.8 slm 20 torr

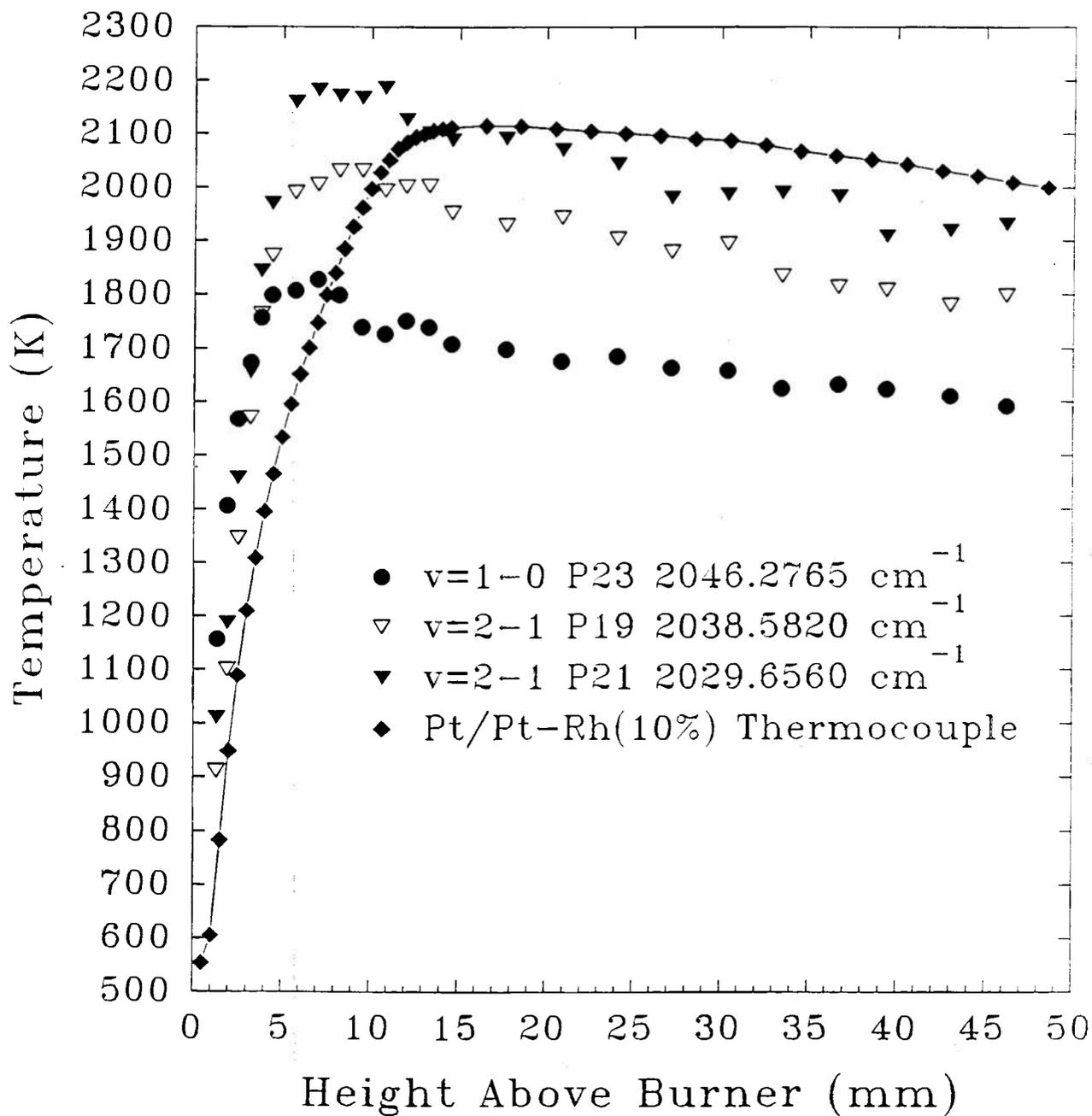


Figure 3. Temperature profile of a 20-torr stoichiometric $C_2H_4/O_2/Ar$ (0.4/1.2/2.8 slm) flame as determined from the Doppler width of the three indicated CO transitions and measured with a Pt/Pt-Rh(10%) coated thermocouple.

than does the thermocouple profile. Since the spatial resolution of the laser beam is 2 mm, the measured Doppler temperature at a given point in the flame is an average of temperature of the region 1 mm above and below the center of the laser beam. At points near the burner surface, in the preheat zone of the flame, the temperature gradient is steeper than in any other region in the flame. Consequently, the 2-mm laser beam samples CO molecules at a large range of temperatures which causes the discrepancies between the measured Doppler and thermocouple temperatures. This effect will be magnified if the infrared beam is not exactly parallel to the burner head, which is believed to be the case during the course of this experiment.

The temperature profile of a 20-torr CH₄/O₂/Ar (0.88/1.76/2.8 slm) flame measured from the Doppler width of the three CO transitions listed in Table 1 are compared to Pt/Pt-Rh(10%) thermocouple measurements in Figure 4. Similar to the C₂H₄/O₂/Ar results, the temperatures determined from the Doppler widths of the two measured hot band transitions compare well with the thermocouple measurements high above the burner surface and the temperatures determined from the fundamental band transition are consistently low. The Doppler temperature profiles also peak closer to the burner surface in relation to the thermocouple profile. These results suggest the Doppler widths of the two CO hot band transitions can yield reliable absolute temperatures in the post flame region of low-pressure flames given that accurate values for the collisional broadening parameters are known. In addition, the agreement between the thermocouple measurements and the temperatures determined from the fundamental band transition may improve with the elimination of the interfering CO absorption which occurs outside the edges of the flame along the line-of-sight. Improved agreement between the tunable diode laser data and the thermocouple measurements can also be achieved by increasing the spatial resolution of the technique with tighter focusing of the infrared beam.

5. SUMMARY AND CONCLUSIONS

A novel flame thermometric technique has been developed which is based on the infrared spectroscopy of flame species as measured with a tunable diode laser. Temperature profiles for 20-torr premixed C₂H₄/O₂/Ar and CH₄/O₂/Ar flames were determined from the Doppler width of individual CO ro-vibrational transitions recorded at various heights above the burner surface. The Doppler temperature profiles compare well with the results obtained from Pt/Pt-Rh(10%) thermocouple measurements. Improving the spatial resolution of the laser beam and determining more accurate values for the collisional broadening parameters for the CO transitions probed in this study will increase the reliability of this

CH₄/O₂/Ar Flame

0.88/1.76/2.8 slm 20 torr

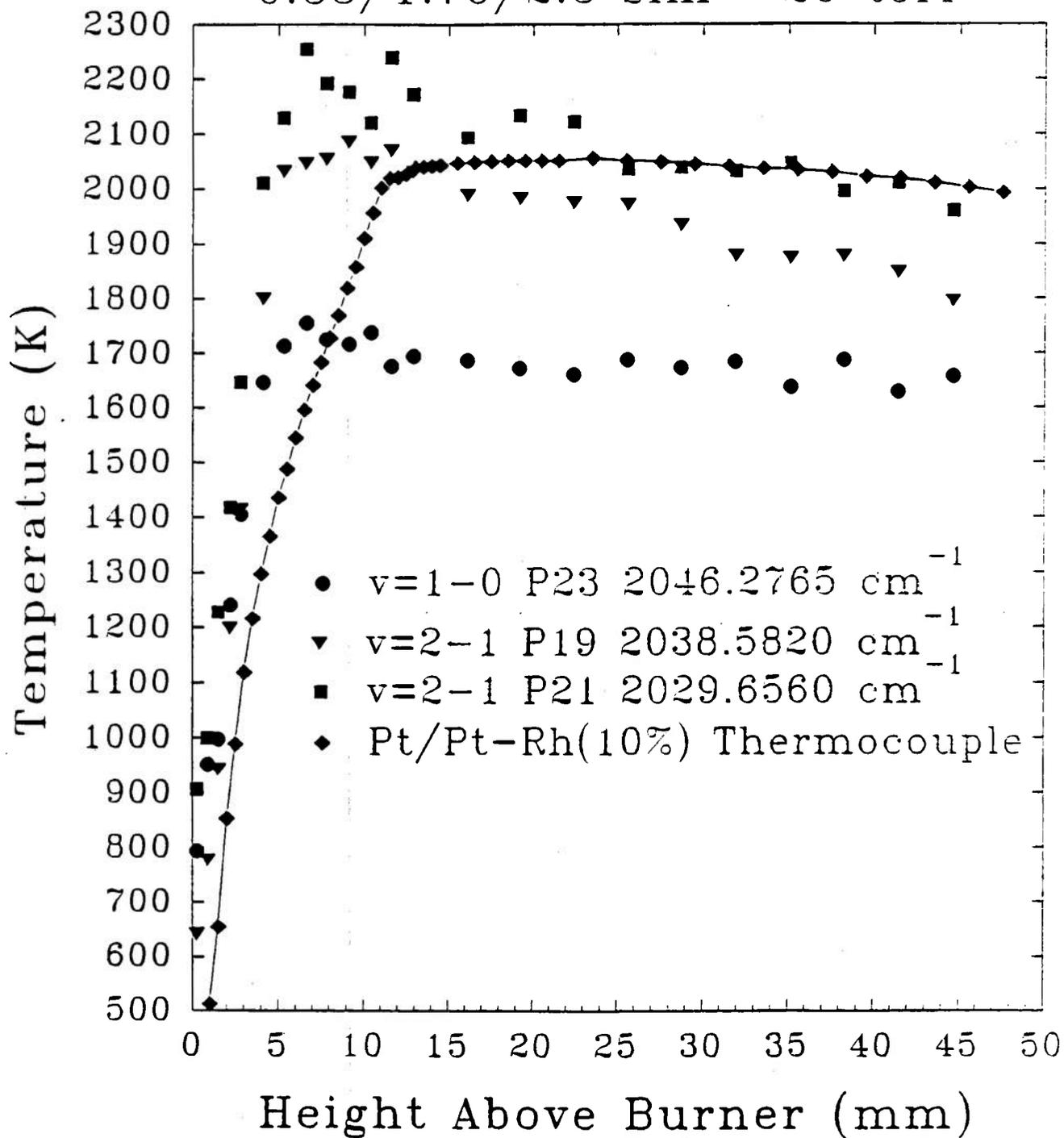


Figure 4. Temperature profile of a 20-torr stoichiometric CH₄/O₂/Ar (0.88/1.76/2.8 slm) flame as determined from the Doppler width of the three indicated CO transitions and measured with a Pt/Pt-Rh(10%) coated thermocouple.

technique and result in more precise measurements of absolute temperature in low-pressure premixed flames. One potential drawback is that the Doppler width varies as a function of $T^{1/2}$, whereas the two-line technique described in the literature varies linearly with temperature. The Doppler temperature measurements, however, could prove to be a more versatile temperature measurement than the traditional two-line method because any CO transition potentially can be used for temperature measurements as opposed to using a pair of CO lines whose transition frequencies are close enough to scan within the confines of a given diode laser mode. In environments where temperatures are considerably higher than in low-pressure flames, for instance low-pressure plasmas, significant spectral overlap between two adjacent ro-vibrational transitions could lead to errors in measured temperatures. This complication can be avoided if the temperature is determined from the Doppler width of a single ro-vibrational transition.

In the future, tunable infrared diode laser spectroscopy can be used to measure absolute CO concentration profiles for comparison with the profiles determined from mass spectrometric detection of CO and those calculated in flame models. This method can readily be extended to the study of NO in propellant-like flames. This will be particularly important in flames in which NO_2 is the primary oxidizer where other laser-based flame diagnostics have proven difficult to implement. In addition, the rapid scanning capabilities of the diode laser (in the MHz range) can be utilized to study time resolved perturbations in flames such as turbulence. Spectral profiles recorded in rapid succession can be used to follow local temperature variation as a function of time.

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