Fast Temperature Determination by Nitrogen Coherent Anti-Stokes Raman Spectroscopy

S. Fujii* and M. Gomi*
National Aerospace Laboratory, Chofu, Tokyo, Japan

A new method of measuring gas temperature by a laser probe, coherent anti-Stokes Raman spectroscopy (CARS), is presented. The ratio of signal intensity from two photomultipliers is used, instead of a multichannel analyzer, for the instantaneous temperature determination on the basis of Q-branch nitrogen molecule spectral profiles. The proposed spectroscopic procedures feature real-time, on-the-spot temperature measurement for a high rate of laser shot repetition using low-cost optical and electronic instruments. Application to burners was conducted with reasonable results.

Nomenclature

c	= speed of light
$\hbar$	= Planck constant
h	= $h/2\pi$
i	= $i^2 = -1$
J	= rotational quantum number
N	= population
T	= absolute temperature
V	= vibrational quantum number
ω	= frequency
ω_1	= pump frequency
ω_2	= Stokes frequency
ω_3	= CARS frequency
χ''	= resonant susceptibility
σ	= standard deviation
Γ_J	= line width at J
ϕ	= equivalence ratio
$(\partial\sigma/\partial\Omega)$	= normal Raman cross section

Introduction

OWING to pioneer work,[1-4] coherent anti-Stokes Raman spectroscopy (CARS) thermometry has been widely applied in combustion diagnostics[5-7] and other fields. Nitrogen molecules are sometimes chosen as CARS probe molecules because of their inertness and their presence in large concentrations in hot and chemically reacting flows. For such large populations, the CARS signal becomes a coherently intense light and is approximately expressed in terms of resonant third-order symmetry susceptibility

$$-3\chi'' i = \sum_J \frac{2Nc^4}{h\omega_2^4}\left(\frac{\partial\sigma}{\partial\Omega}\right)_J T_J \Delta_J \qquad (1)$$

when the phase match condition $\omega_3 = 2\omega_1 - \omega_2$ is satisfied. That is, the nonresonant electronic term also contributes to the CARS signal, but can be ignored when the resonant Raman contributions are large. If the vibrational levels are in equilibrium at temperatures T, the Q-branch population difference, ΔN_J, between V and $V+1$ at J depends upon the absolute temperature T according to the Maxwell-Boltzman distribution functions; that is, the normalized profile of susceptibility vs J is a function of temperature only. The comparison of spectral profiles between experiment and theory then allows deduction of the temperature.

CARS has many advantages in its application, in particular, excellent discrimination against luminous or fluorescent backgrounds. However, the present authors, during a series of practical applications, have found some drawbacks. Most of the previous measurements have been carried out by multiplex CARS where the broadband ω_2 is employed. The acquisition and storage of broadband spectral profiles may need a photodetector and analyzer with multichannels and a monochromator with significantly high resolution. Such devices are expensive and, generally a long time is required to sweep the whole spectral range or execute partial integration[8] for fitting, using a medium-sized computer. Without feeding the measured data directly into a large-scale computer, therefore, it is not possible to acquire high-repetition temperature information on the spot.

The present paper presents a new method to simplify the spectroscopic procedures, which results in real-time determination of temperature with a remarkable decrease in optical and electronic instrumentation costs.

Principle of Fast Temperature Determination

Figure 1 shows an experimental setup consisting of two signal channels. One is a monochromator-detector-OMA channel and the other a monochromator-two photomultiplier-S/H channel. The 700 mJ/pulse Nd:YAG laser (Molectron model 34-20) with 20–30 Hz repetition, homemade dye cell of 45% conversion efficiency, flat flame burner, and computer were common to both. The detector-OMA channel provided the 400 multichannel data points in a nitrogen Q-branch spectral domain, as is shown in Fig. 2. The theory/experiment curve fitting determined a flame temperature of 2002 K with an uncertainty of ±25. This procedure has been in general use for precise temperature determinations, but the work of curve fitting is sometimes troublesome. As one alternative method, the present authors[5] reported that the ratio of hot to cold peak intensities gave the temperature but with somewhat large errors of ±50 K. The large uncertainties result from the sensitve peak positions. The method of taking the total area under the peaks much improves the measurement precision. Along this line, Eckbreth et al.[8] used the integral intensity of warm, hot, and cold bands to evaluate temperature in their seven methods. However, they had to calculate numerically the area using the spectrum already stored in the computer for each laser shot. As the repetition rate of laser shots increases, therefore, the number of data points increases tremendously, making the method unlikely to be able to fur-

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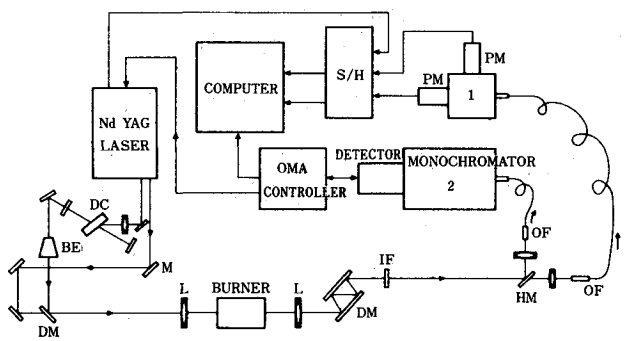
*Noise and Emission Research Group.

nish instantaneous time-varying temperature on a real-time basis. To improve this situation, our new spectroscopic technique uses the two photomultiplier-S/H channels as shown in Fig. 1 to obtain profile areas directly from the light intensity as it passes a predetermined mechanical slit width, without recourse to numerical integation.

The #1 monochromator had a resolution of 0.03 nm at FWHM. In the #1 monochromator in Fig. 3, a half-silvered mirror was positioned to divide the dispersed signal light onto two slits, each viewed by a photomultiplier. It is clear that the ratio of the two signal intensities should depend strongly on the sample gas temperature. To confirm this point, parallel data acquisitions were performed by use of the two channels in Fig. 1 for a laminar flame. The data of an ensemble average of over 100 shots were first plotted by the rotation of a reflective grating of the #1 monochromator for a narrowband CARS, as shown in Fig. 4. The flame temperature at that point was determined by full fittings in the OMA channel with a high-resolution monochromator. Such procedures revealed the characteristics and instrument function of the #1 monochromator in detail. By use of the known temperature and the measured/theoretical spectrum, the slit function of the #1 monochromator was determined as 5.5 cm^{-1}. By use of the obtained slit function at the reference point, the area at the other temperature was theoretically obtained from the numerical integation on the Q-branch spectrum. The theoretical profiles at three temperatures and the relationship of the two slit widths are shown in Fig. 5. The ratio of the one area produced by the #2 slit width to the other area of #1 was then calculated and plotted against temperature in Fig. 6.

To observe the slit sensitivity, two initial settings of the #1 slit width were selected: 2112.5 cm^{-1}, denoted as Case 1, and 21124.5 cm^{-1}, Case 2. At this stage, the hardware of Fig. 1 provided extensive and rigorous comparisons of measured temperature using multichannel full fitting and two-photomultiplier intensity ratios, respectively, at the same points in space and in time for the laminar flames, as shown in Fig. 7. A 100-shot ensemble average was taken for all measurements. The data scatter inherent in the CARS system at room temperature is shown in Fig. 8. Note the significantly smaller rms value obtained with the photomultiplier intensity-



DC: DYE CELL, BE: BEAM EXPANDER, DM: DICHROIC MIRROR, OF: OPTICAL FIBER, HM: HALF MIRROR, IF: INTERFERENCE FILTER, S/H: SAMPLE HOLDER, OMA: OPTICAL MULTICHANNEL ANALYZER

Fig. 1 Experimental setup.

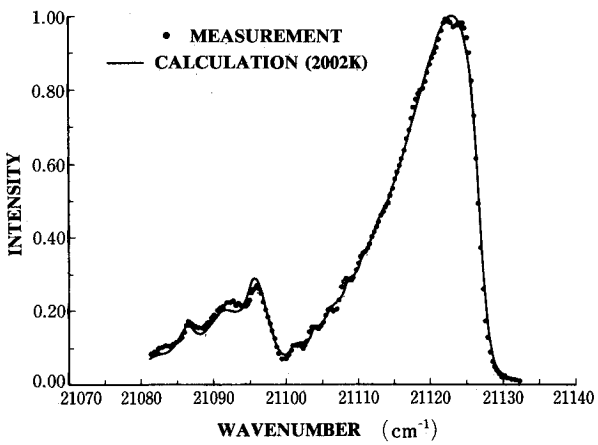


Fig. 2 Nitrogen Q-branch spectrum at high temperature.

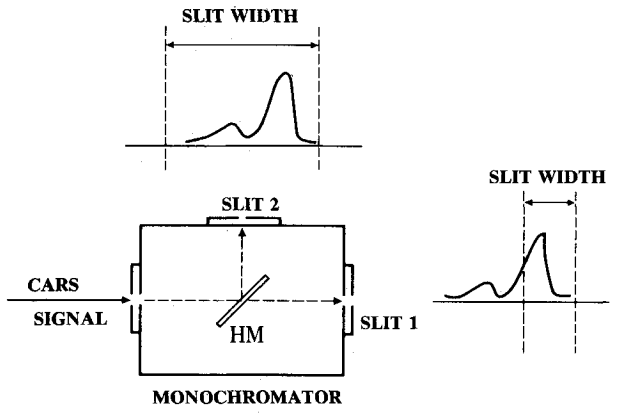


Fig. 3 Relation of monochromator and slit 1 and 2.

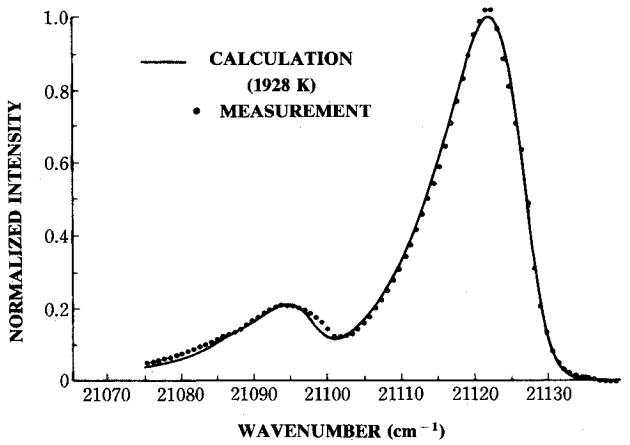


Fig. 4 Curve fitting with experimental data at the reference point ($T = 1928 \pm 24$ K.)

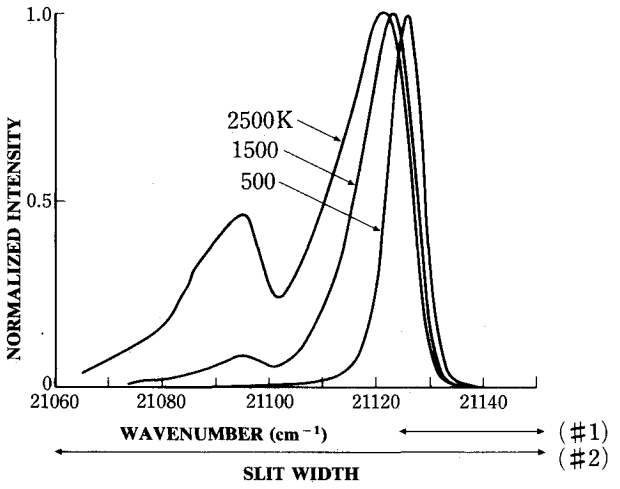


Fig. 5 Slit widths in theoretical spectrum.

ratio method as compared to that with the OMA full-fitting determination. The straight line relationship, displayed in Fig. 7, demonstrates the validity of the methodology, which compared cold-band intensity to the whole sum of the cold and hot bands. It should be noted that the present method has the capability of determining only vibrational temperature; therefore, its application to a system, where equilibrium between the rotational and vibrational degrees of freedom cannot be assumed, is questionable.

Application to Burners

A Bunsen burner used in the previous study[5] was again selected to investigate further the characteristics of the present method. A mismatched optical alignment[9] was applied with a focal length of 100 mm. The mean temperature averaged over 1000 events and its histogram are shown in Figs. 9 and 10. The measured temperature showed a steep gradient near the burner edge. The histogram broadening involves errors introduced in the whole system of measurement as well as turbulence fluctuations. Note that invalid data outside of $\pm 3\sigma$ were rejected. The value of rms dispersion was maximized near $R=6$ mm where the mean temperature assumed the steepest slope.

As implied in Eq. (1), the signal level depends on the second power of the population, N^2; further, the hotter part of the whole volume emits a stronger intensity than the remaining colder part in high-gradient temperature fields. With this in

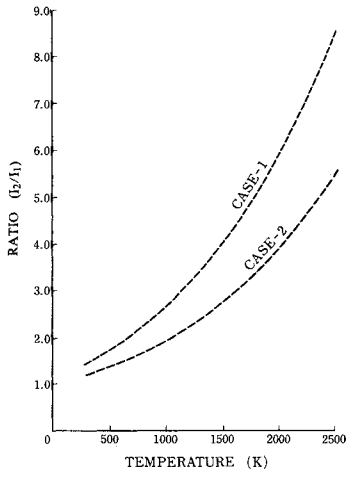


Fig. 6 Theoretical curve of photomultiplier intensity ratios.

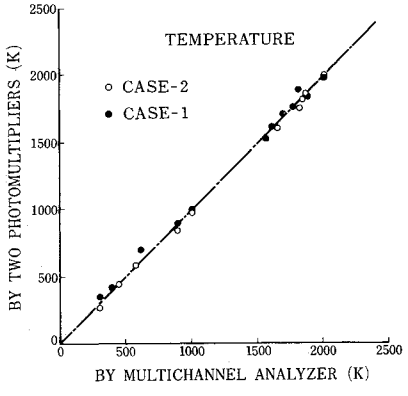


Fig. 7 Comparison of two temperature measurements.

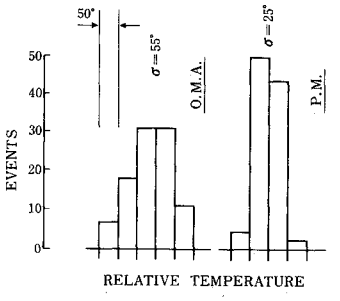


Fig. 8 Room temperature measurements.

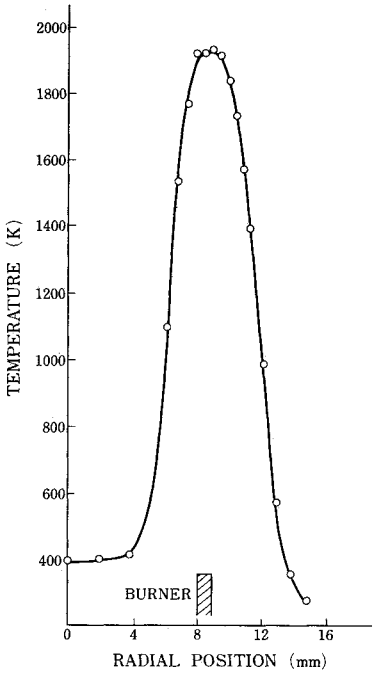


Fig. 9 Measured temperatures.

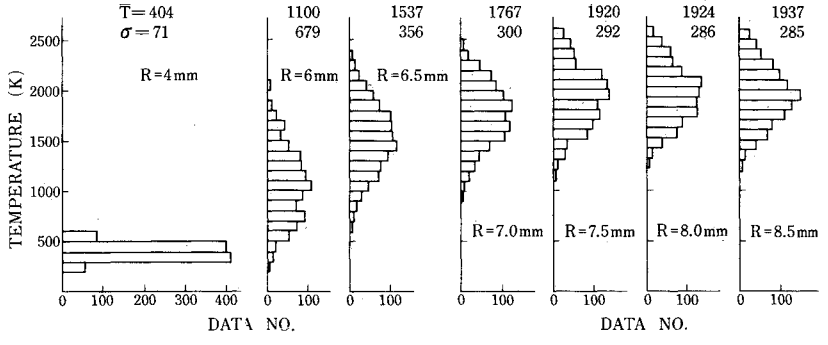


Fig. 10 Temperature historgrams for Bunsen burner.

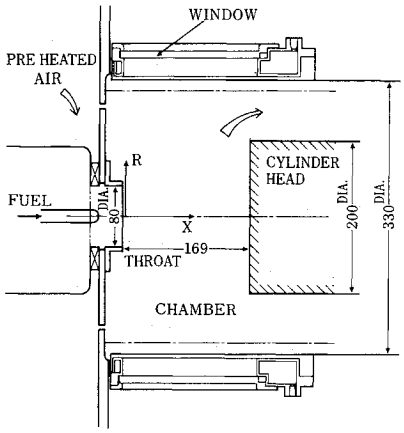


Fig. 11 Large-scale burner.

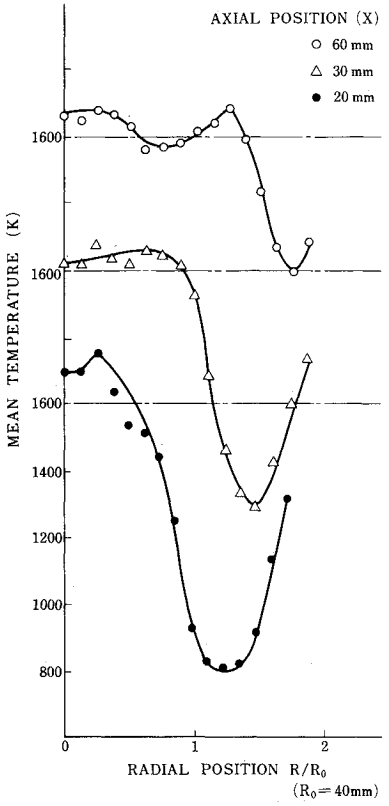


Fig. 12 Radial temperature distribution ($R_0 = 40$ mm).

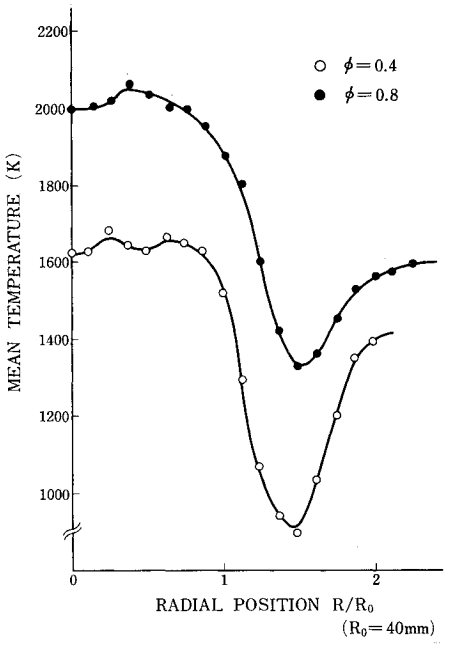


Fig. 13 Change of temperature patterns by equivalence ratio (ϕ).

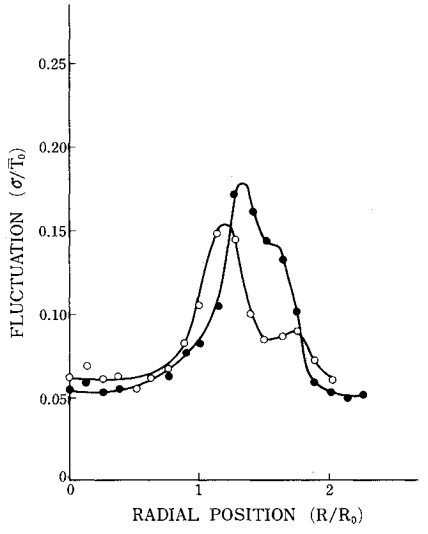


Fig. 14 Temperature historgrams.

mind, a rigorous test was attempted by slowly moving the phase match volume in a normal direction of the circle burner from the low to high temperature points. When the sample volume was well within the zero-gradient low-temperature region, the histogram obtainable from 1000 shots exhibited a near-Gaussian distribution. When the volume edge touched on a high-gradient region, the histogram height indicating a maximum temperature group gradually increased. As the volume moved further, such disturbance shifted onto the minimum cold side of the histogram and finally disappeared.

The traveling distance between disturbance incipiency and return to near Gaussian might represent a spatial resolution for steep gradient fields. Such repeated attempts showed that the sample volume could be evaluated as 2.0 mm (length) × 10 μm (diam) at maximum. For zero or small gradients, the effective length decreased down to 1.0 mm.

A 330 mm diam combustor was next used for fast CARS measurement as noted in Fig. 11. Air preheated to 800 K was swirled in and mixed with propane gas to produce a turbulent diffusion flame near the throat. A thermally coated body simulating a cylinder head was located in the downstream region (because the original purpose of the hardware lay in research of a Stirling engine burner). Quartz window slits provided access to the laser beam passes. An Nd:YAG laser and power units were enclosed in an air-conditioned booth close to the burner. The received CARS signal was piped by an optical fiber of 400 μm core diam to a data processing room where the monochromator, photomultipliers, and computer were positioned. Voltages from the photomultipliers were raised by a high-response amplifier. The time of data reading was controlled by a sample-hold mechanism in which a time lag was set up at the Q-switch start of laser beam emission.

Measured data are summarized in Figs. 12-14. All data of 1000 events at one measuring point were screened by $\pm 3\sigma$ criterion. A focal length of 500 mm was needed to prevent heat damage to the lenses. The spatial resolution was estimated as 10 mm (length) × 50 μm (minimum diam) for high-gradient measurements. The axial resolution might be improved up to 5 mm at most in near-flat regions. The mean temperatures were sensitive to the change in equivalence

ratios, which represent the fuel/air mixture ratio normalized by the stoichiometric value. Again, the temperature fluctuations were maximized in the steep gradient zone of mean temperature shown in Figs. 13 and 14. However, such fluctuations contained two components. One was the fluid mechanical turbulence in the flame and the other measurement errors. The histogram spread from noise might result from the dye qualities of the dye laser beam, the monochromator, or other sources. Beam quality scrutiny is needed on these points in future research.

Conclusions

A spectroscopic technique for CARS temperature measurements is proposed herein. The comparisons between the present method and the conventional multichannel analyzer showed little difference in the mean temperature. Data scatter introduced in the measurement system was much smaller with the photomultipliers than with the multichannel at room temperature.

Using the proposed method, instrument costs can be significantly decreased, that is, a lower-resolution monochromator, and more sensitive but less costly photomultipliers than those used in the conventional multichannel method can be employed. It was confirmed that the ratio of the two photomultiplier intensities gave instantaneous temperature information on a real-time basis for a high rate of laser shot repetition. Application of the CARS system to small and 360 mm diam burners was successfully achieved.

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