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A SIMPLIFIED CARS MEASUREMENT SYSTEM FOR RAPID DETERMINATION OF TEMPERATURE AND OXYGEN CONCENTRATION

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SUMMARY

A new spectroscopic concept for the rapid determination of temperature and oxygen concentration by CARS (Coherent Anti-Stokes Raman Spectroscopy) was described. The ratio of two spectral regions in the broadband Q-branch spectrum was detected by photomultipliers in a monochromator, which ratio depends on temperature and species concentration. The comparison of the measured data with theory was made using a flat flame burner and an electric furnace, with reasonable results. Various optical techniques for alignment were introduced including a highly efficient, stable dye oscillator. The combination of the spectroscopic concept and the optical techniques will make the CARS measurement system rapid in data processing and simple in optical parts.

NOMENCLATURE

T	gas temperature, in Kelvin
$\chi^{(3)}$	third-order susceptibility
χ'	unsymmetrical term
χ''	symmetrical term
χ_{NR}	nonresonant term
O_2	angle between the pump and Stokes beam
ΔW	separation distance between two hemicircle beams
ω_1	pump frequency
ω_2	Stokes frequency
ω_3	CARS frequency

$i^2 = -1$

INTRODUCTION

CARS (Coherent Anti-Stokes Raman Spectroscopy) techniques have been widely applied in combustion diagnostics. For CARS thermometry, nitrogen molecules are often chosen because of their large concentrations available as an inert gas. In combustion gases, the concentration of diatomic molecules such as O_2 and CO may be easily determined by CARS because of their simple theoretical spectrum.

Most measurements have been carried out by multiplex CARS where the broadband of ω_2 is employed. CARS has many advantages in its application. It has excellent discrimination against luminous or fluorescence backgrounds. In contrast, the present author has found a few drawbacks during a series of practical applications. By use of one optical multichannel analyzer, it takes appreciable time to record the intensity in multichannels and then compare the measured data with theory. As the repetition rate of the laser increases, it will be unable to furnish the instantaneous time-varying temperature and concentration on a real-time basis. The acquisition and storage of broadband spectral profiles may need a photodetector and analyzer with many channels and a monochromator with significantly high resolution. Such devices are quite expensive. This paper proposes a new method to simplify the spectroscopic procedures, which results in real-time determinations of temperature and concentration. Various optical techniques are also introduced. The combinations of the new spectroscopic concept and these new optical techniques will make the CARS system fast in data processing and simple in optics, with a remarkable decrease in optical and electronic instruments costs.

RAPID TEMPERATURE DETERMINATION BY NITROGEN CARS

The CARS intensity depends on the squared third-order susceptibility of molecules in the gas, which can be written as

$$|\chi^{(3)}|^2 = |\chi' + i\chi'' + \chi_{NR}|^2 \qquad (1)$$

Nitrogen molecules are often chosen for CARS temperature measurements because of their large concentrations in hot and chemically reacting flows. For such large populations, the resonant symmetric term $|\chi''|^2$ is dominant and other terms $|\chi'|^2$, χ_{NR}^2 and $2|\chi'|\chi_{NR}$ are negligible. The spectral profiles of $|\chi''|^2$ depends only on temperature through the Maxwell-Boltzman equilibrium distribution for the Q-branch spectrum. The comparison of experimental profiles with theory gives information on temperature. However, it takes some time to sweep the voltages of intensity in the multichannels and then compare the measured data with theory in the whole Q-branch range. Because of the amount of time necessary to do this comparison, real-time measurements are not possible.

Instead of using the whole Q-branch spectrum, the ratio of partial integrals such as hot/cold bands, warm/cold bands and their combinations have been reported[1]. However, the integrations were performed numerically on the measured data after the optical multichannel processes, which still did not give the real-time data reduction. The peak ratio of hot/cold bands might provide one possibility for speeding up the process but with rather large errors[2]. As another alternative, a new spectroscopic concept is proposed herein.

The basic idea starts by splitting the CARS signal using spectrometer exit slits #1 and #2, as shown in figure 1. By changing the two slit widths, various intensity ratios of slit #1 to #2 could be optically obtained.

A propane-air mixed flat flame burner was employed in the experiment. By use of the burner, a nitrogen spectrum at high temperature was obtained by scanning the monochromator. The results are shown in figure 2. We made an average of over 100 pulses. The best fit curve was obtained by adjusting the value of the slit function in the computer code, as indicated in figure 2. The value obtained from the best fit was 5.5 cm^{-1}. The spectra at three temperatures with this slit function were convoluted in figure 3, where the widths of slits #1 and #2 are also illustrated along the horizontal axis. The intensity ratio for slit #2 to slit #1 was calculated in the temperature range from 300K to 2500K and plotted in figure 4 and is denoted by CASE(1). The width of slit #1 was selected to give the curve sensitivity to the temperature change. To see the effect of slit width, the width of slit #1 was slightly widened. The result is drawn in figure 4 as CASE(2).

To compare the present method with the conventional whole-spectrum method, simultaneous measurements were performed. The test setup shown in figure 5 consisted of two optical paths. The 700mJ/pulse Nd:YAG laser with 20Hz repetition rate, homemade dye oscillator, flat flame burner and computer were common to both paths. The CARS signal was generated within the burner and split by a half mirror. One of the CARS signals was passed through the monochromator with a half mirror inserted, detected by a photomultiplier followed by a sample-hold circuit and then fed to the computer. This is the path for the new concept. The other signal was processed by the monochromator, silicon-intensified array detector and optical multichannel analyzer. The latter describes the lineup for a standard CARS measurement. It is further noted that the monochromator associated with the two photomultipliers had much less resolution (0.03nm, FWHM), compared to that (0.007nm) of the monochromator used in the conventional arrangement.

Extensive and rigorous comparisons were made for the measured temperatures by the conventional CARS, and by the two-photomultiplier method. The results are summarized in figure 6. The straight line relationship validates the new method. It was not sensitive to the change of slit widths within the data range of CASE(1) and CASE(2). Note that all data were obtained from the average over 100 pulses.

RAPID CONCENTRATION DETERMINATION BY OXYGEN CARS

In a combustion gas, oxygen is sometimes detected in low concentrations because it is reactive. In such a situation, the two terms of $|\chi'|^2$ and $|\chi''|^2$ are neglected and the susceptibility can be written as,

$$|\chi^{(3)}|^2 \sim \chi_{NR}^2 + 2|\chi'|\chi_{NR} \qquad (2)$$

Equation (2) indicates a significant domination of the nonresonant term χ_{NR}, which implies a difficulty in detecting low concentrations. One method of overcoming this difficulty is to use a polarization technique for rejecting χ_{NR}. However, the disadvantages in using polarized CARS are twofold. One is associated with the considerable attenuation of signals under the conditions of extremely low signal-to-noise ratio. The other is that the measurement system is further complicated by the addition of expensive polarization devices. Instead of rejecting the nonresonant term, the intensity ratio method was again employed.

Figure 7 shows a typical computed O_2 spectrum of the Q-branch at a concentration of 3% in volume for a temperature of 1500K. The same slit function of $5.5cm^{-1}$ as that obtained in the temperature experiment was used with a nonresonant susceptibility of $10.97 \times 10^{-18} \times (273.15/T) cm^3/erg$. The susceptibility given by equation (2) depends on both temperature and concentration. We first calculated the ratio of numerical integrals #2/#1 and next those #3/#1 at various temperatures and concentrations. The results are plotted in figures 8 and 9. The integral over #1 represents the magnitude of χ_{NR}. Therefore, the integral ratio shown in figure 8 can be considered the hot/nonresonant ratio, which is rather insensitive to the change of temperature beyond 1200K but still sensitive to the concentration variation. On the other hand, the ratio of #3 to #1 in figure 9 is the warm/resonant ratio and is useful for concentration determinations below 1200K.

A small electric furnace was used to compare the measured and calculated ratios and is illustrated in figure 10. Using pure nitrogen gas, we first made the CARS temperature measurement within the furnace and determined the relationship between the gas temperature and the wall thermocouple prior to all tests. The uniformity of gas temperature within the furnace space was also ascertained. In the oxygen detection tests, the O_2 gas, diluted by N_2, was introduced into the furnace and heated up to the predetermined temperature. At an O_2 concentration of 8%, the data were obtained in the temperature range of 300K up to 900K at 100K steps. At 4%, three data points at 300K, 600K and 900K were collected. Data of 1% concentration were obtained only near room temperature (300K) and the maximum temperature of 900K. The measured data are plotted in figure 9. All data were obtained from the 100 pulse ensemble average. The agreement with theory was reasonable. It was not possible to provide measured data beyond 900K due to the furnace capability.

It is further noted that the present method of determining temperature and concentration assumes an equilibrium state between the rotational and vibrational degrees of freedom in the gas to be measured. The application to turbulent flames would be the next step including a rigorous evaluation of measurement uncertainty.

OPTICAL ALIGNMENT TECHNIQUES

MAC Alignment

Various methods have been proposed to accomplish CARS phase matching. The collinear method is popular and easy to implement but not adequate for obtaining high spatial resolution. Although BOXCARS[3] provides for high resolution, it takes some time to realize the precise alignment in a practical large-scale burner. USED CARS[1] may be a useful method for obtaining high resolution but not in general use because it requires the unstable resonant cavity at the laser source to produce a doughnut-shaped beam.

As another method, MAC (Minimum Angle Crossed-beam) CARS is proposed in figure 11. The hemicircles of ω_1 and ω_2, which were made by knife edge insertion, were focused and overlapped only in a small area. In principle, the present method would be a kind of misalignment. However, the overlap was primarily made in the high intensity beam center to avoid the rapid decrease of signal levels due to mismatch.

A small Bunsen burner was used to determine the resolution characteristics of the MAC method. There were two ways to determine the temperature field. One was to make the beams tangent to the burner perimeter and the other to do it normally. The measured temperatures were compared in figure 12 for the different beam separations in both tangential and normal modes. The beam separation of 0.2mm in the tangential mode and 0.6mm in the normal mode gave almost the same temperature profiles along the burner center-to-outside. The small separation of 0.3mm revealed lower temperatures in the high gradient region, which reflected the poor spatial resolution and that large portions of the signal might be from the colder spots. Zero beam separation gave an inaccurate result much influenced by the colder gas.

New Beam Splitting Method

In the CARS measurement, a split of the ω_1 beam is sometimes required. It is proposed that a prism be used to separate the beam into two hemicircles, as illustrated in figure 13. It is of interest to note that the laser beam ω_2 appeared as a hemicircle as it was generated in the dye cell with the hemicircle pump beam. This fact is advantageous for the MAC alignment using the hemicircles of ω_1 and ω_2. One more feature is that the transverse movement of the prism will change the beam split ratio. In other words, the intensity ratio of ω_2 to ω_1 can be varied arbitrarily to obtain the optimum signal, which would be particularly advantageous under adverse environments.

High Efficiency Dye Oscillator

It is important to have a dye oscillator with a high conversion efficiency in the simplified CARS measurement system, otherwise excess laser power as well as dye amplifier would be required. The stability of the laser intensity vs. wavelength is another important factor, in broadband operation, to avoid a reference cell for instability compensation.

Figure 14 illustrates the geometrical relationships for the dye laser emission. Our experience has shown that the angle Θ_2 between the pump and Stokes beams has an effect on the intensity and center-frequency shift for Rhodamine 610, shown in figure 15. Note that the conversion efficiency reached a maximum of 45%. The resonant distance L_2 also was an effective parameter, as shown in figure 16. Besides the geometrical parameters, the concentrations of dye solution and of N_aOH, an additive to the solution, have some effects on the dye lasing characteristics (figures 17 and 18). The output profile is shown in figure 19 for Rhodamine 640. Due to the grating dispersion, it was not possible to obtain data in more than 1nm range at one time. A multiplex CARS generally uses less than 2nm of the Stokes-beam range. Our homemade dye oscillator was stable, flat, and highly efficient over this range.

SIMPLIFIED CARS MEASUREMENT SYSTEM

In figure 20, a simplified N_2 CARS temperature measurement system is illustrated. The laser beam at 532nm was split by the prism. The dye laser output at position B was almost a hemicircle beam whose upper part was cut off by a knife edge in front of position C. At position D, the overlap of ω_1 and ω_2 was done in the lower part of the hemicircle. This is a variation of the MAC method and the signal was generated in the lower part of position E. The laser beams at position F were multireflected on the two opposite sides of the dichroic mirror, which had 80% transmission at 532nm and 97% reflection at 473nm. The interesting feature of the present MAC variant is that the spatial resolution may be adjusted and changed by sliding the knife edge position in front of position F. If the lower part is more cut off, the resolution becomes fine but with a lower signal-to-noise ratio.

In the case of detecting oxygen, we need another set of optical parts to obtain temperature and concentration at the same time, and the focal and collective lens can be in common with the temperature measurement system.

CONCLUDING REMARKS

A spectroscopic concept for the rapid determination of temperature and oxygen concentration has been discussed. The ratio of two intensity integrals in the Q-branch spectrum was employed and the calculated results were compared with the measured data from a flat flame burner and an electric furnace.

A new geometrical alignment, dubbed MAC CARS, was introduced with experimental data from a Bunsen burner. The two hemicircles of ω_1 and ω_2 were overlapped only in the small portion. Large beam separation gives rise to fine spatial resolution. With a small beam separation, the larger contributions to the signal were from the colder spots due to larger axial extents of the sampling volume in the high temperature gradient.

The use of a prism in separating the pump beam might decrease the number of lenses and mirrors necessary for the CARS optical alignment and provide the adequate character in the beam shape for the MAC method.

An example has been designed and proposed for the optical alignment part of the simplified CARS measurement system, where the MAC variant was used. Two sets of such a system will be required to make the simultaneous measurement of temperature and oxygen concentration at the same point in space, with the common focal and collective lens.

It is further necessary that when such systems are applied to turbulent flames, the measurement errors will have to be examined carefully.

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Most of the experiments cited were performed by Mitsuo Gomi at the National Aerospace Laboratory, Tokyo.

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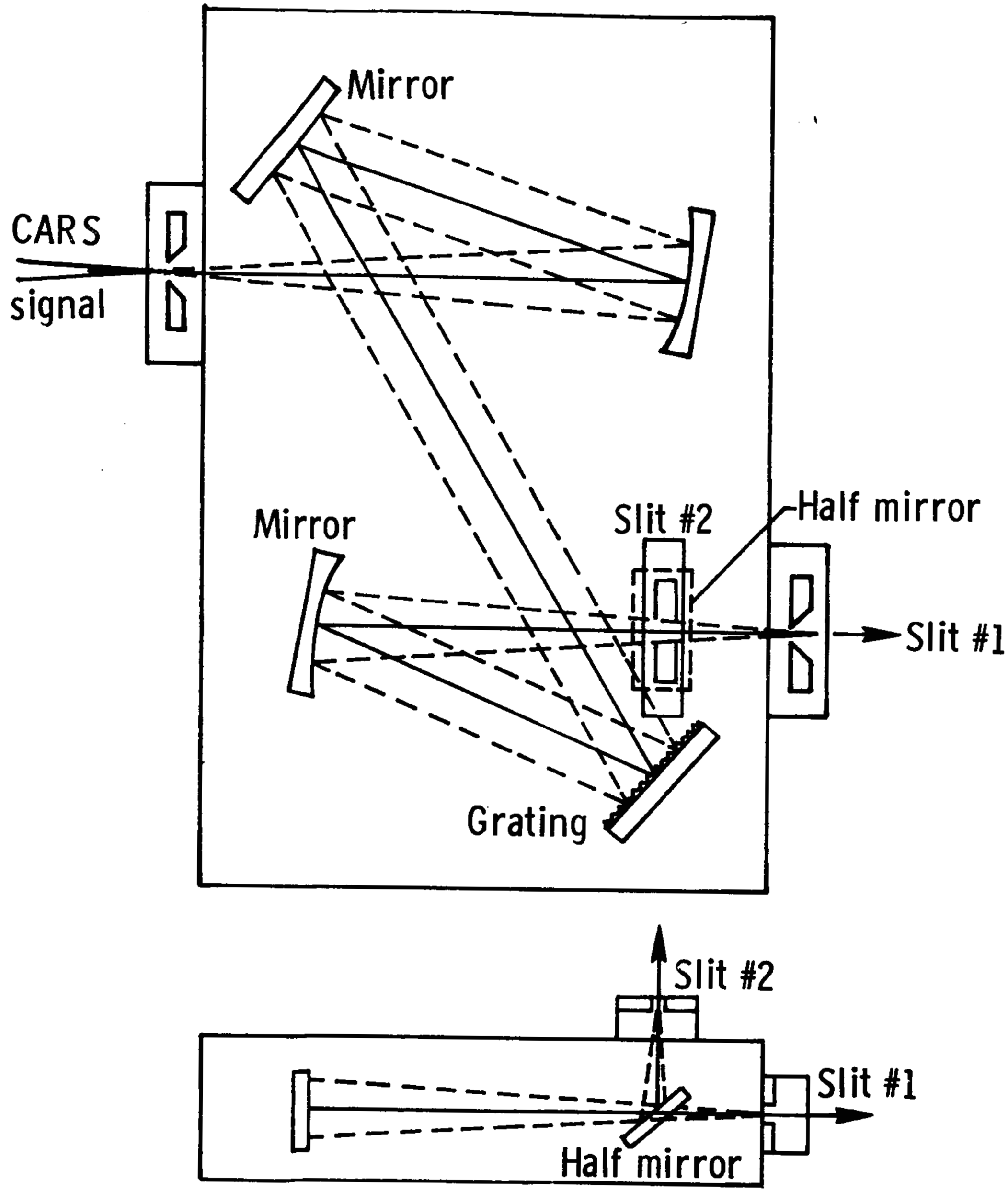


Fig. 1 Monochromator with half mirror inserted

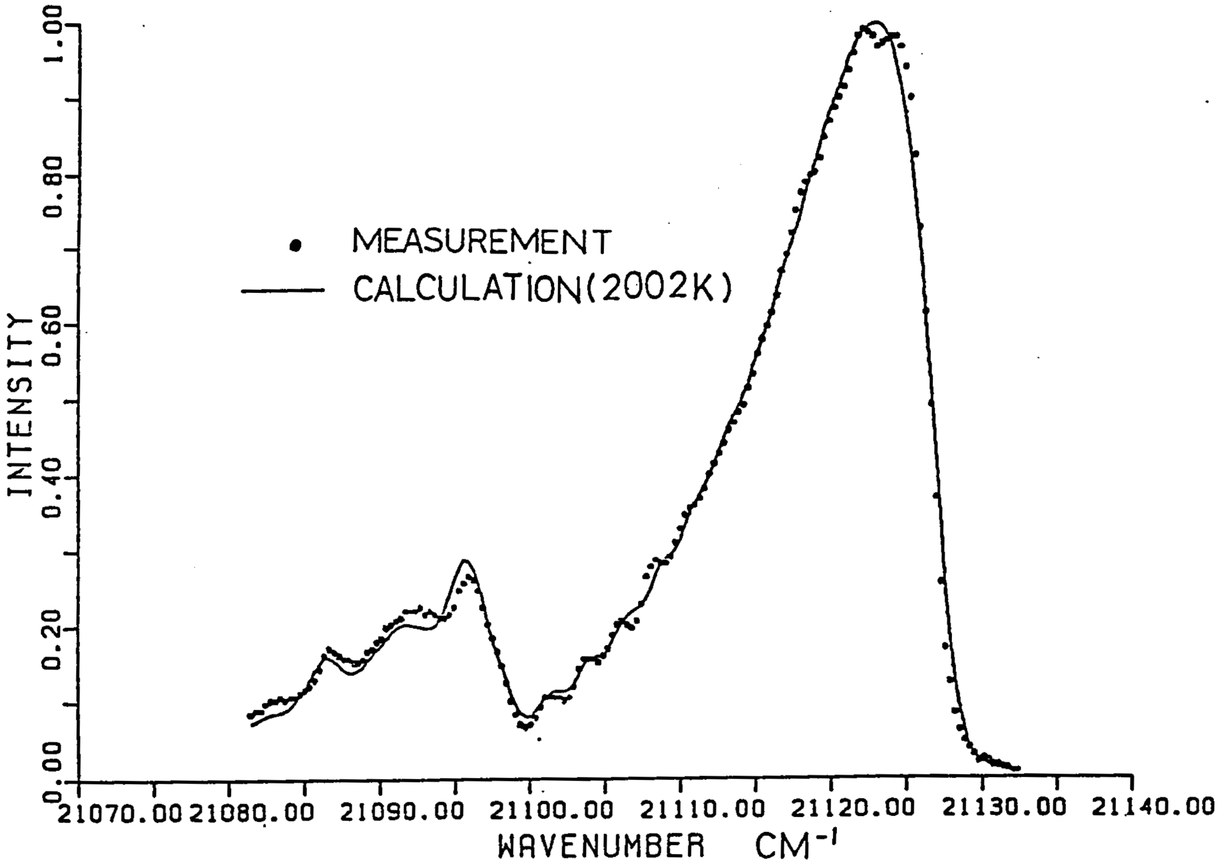


Fig. 2 Nitrogen Q-branch spectrum at high temperature

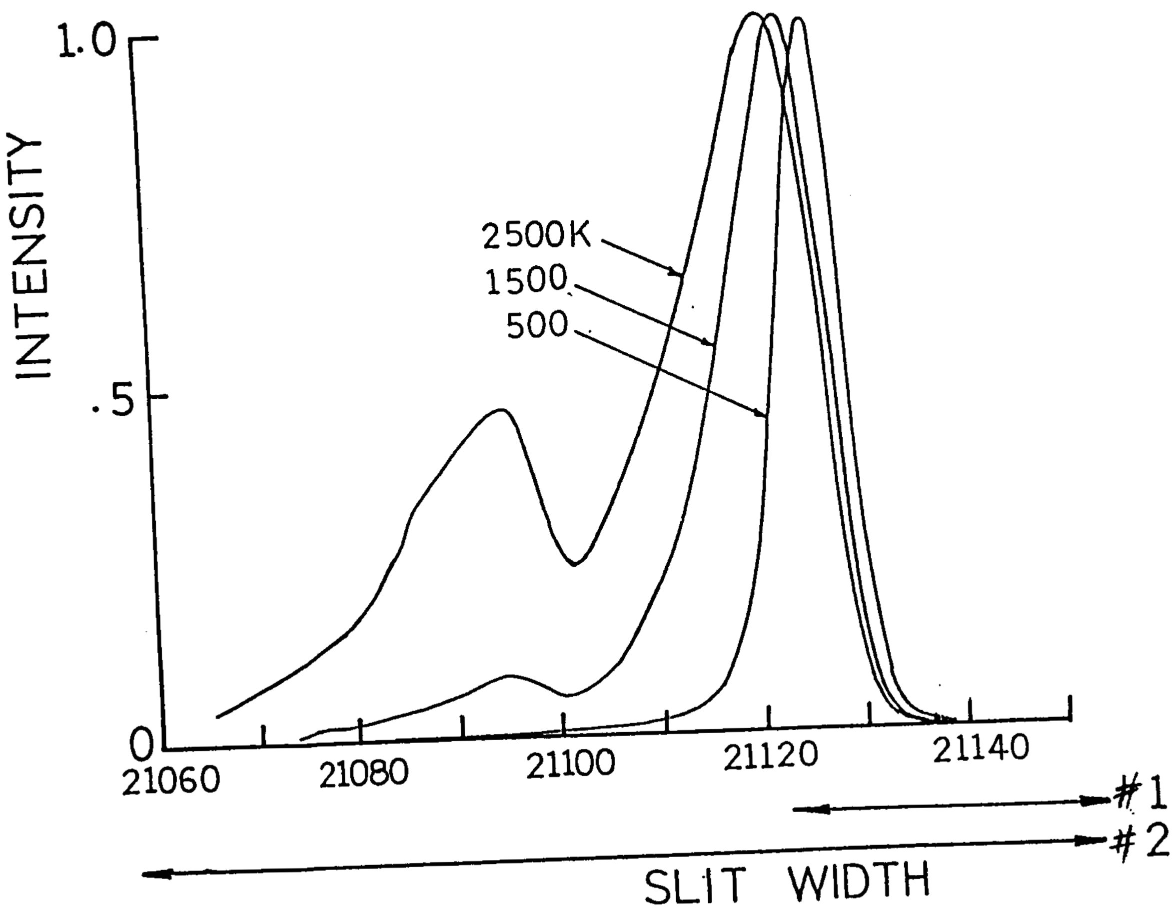


Fig. 3 Calculated spectrum and relationship between slit widths #1 and #2;

#1 = 21122.5 ~ 21155 cm-1

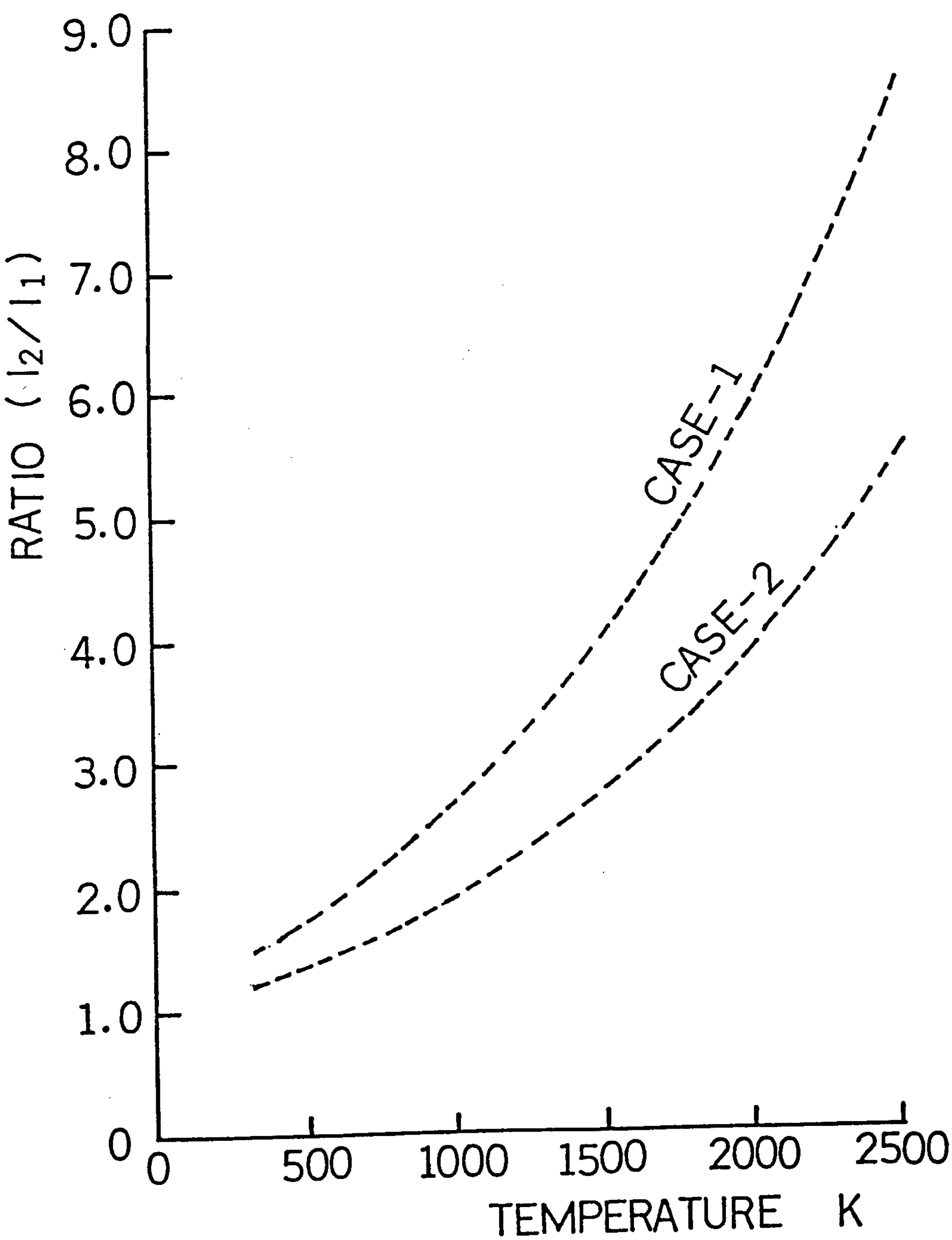
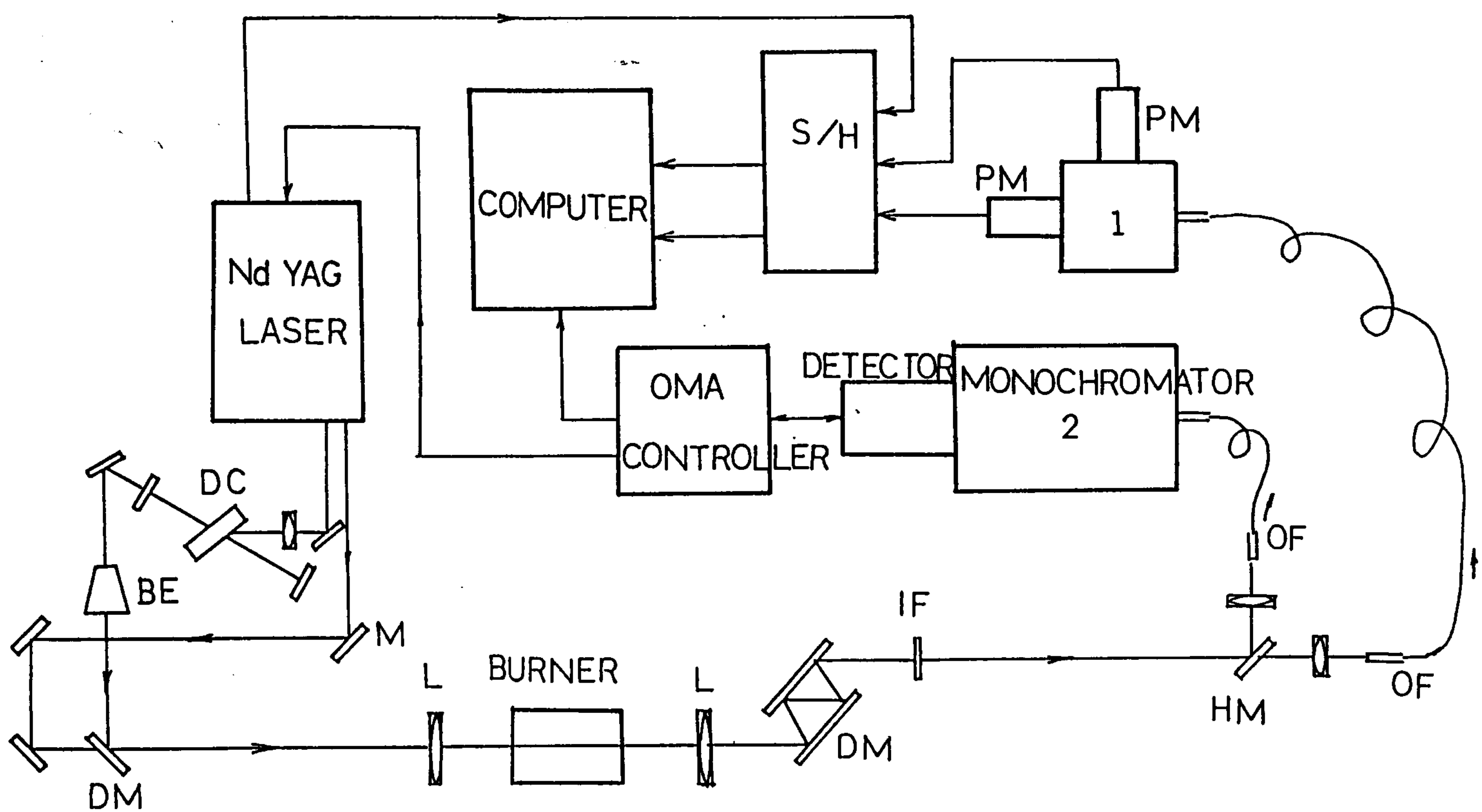


Fig. 4 Calculated intensity ratio; for CASE-2, slit #1 was increased by 2cm-1



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. 5 Experimental setup for parallel data acquisition by conventional and proposed methods

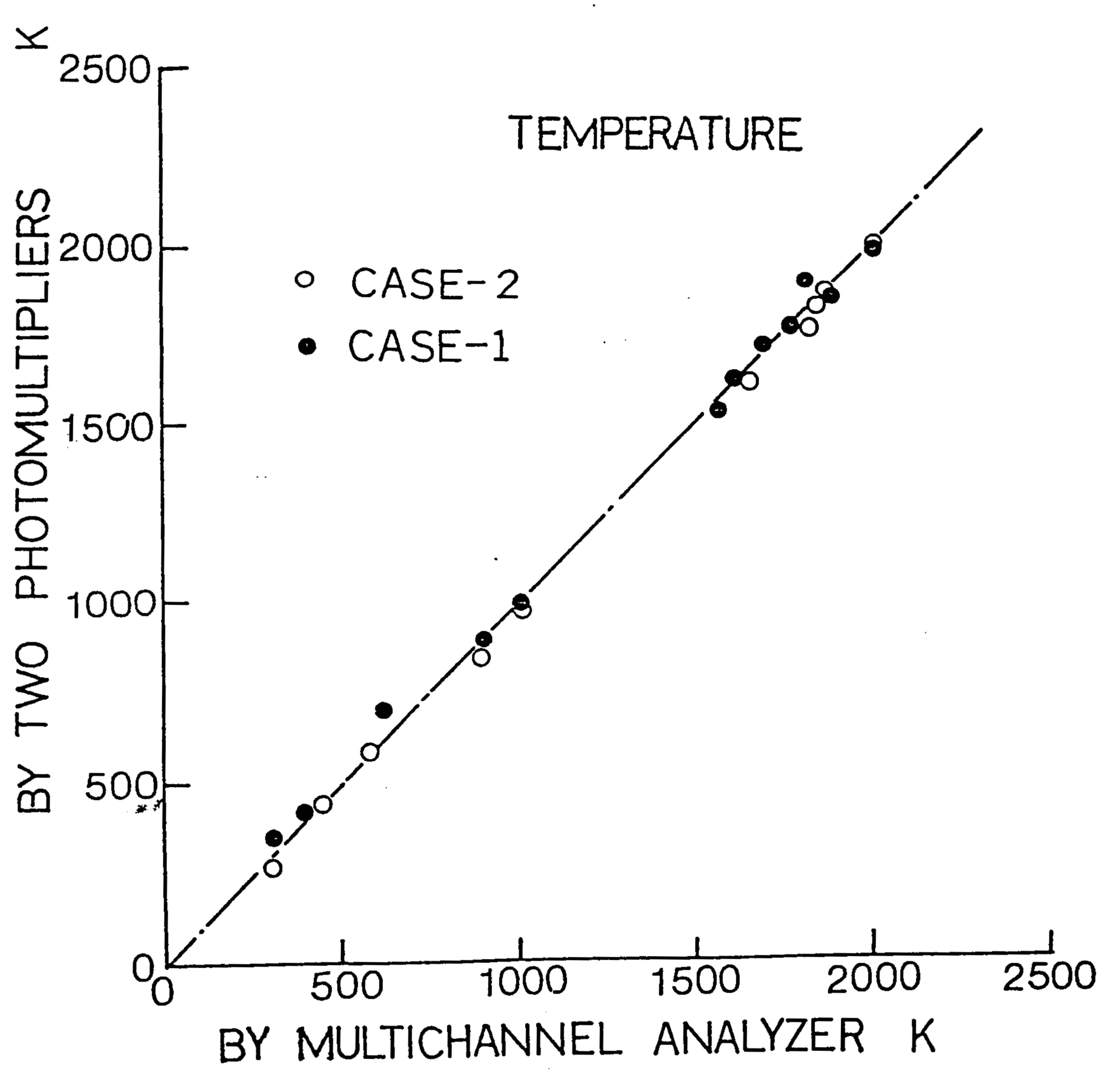


Fig. 6 Comparison of two temperature measurements

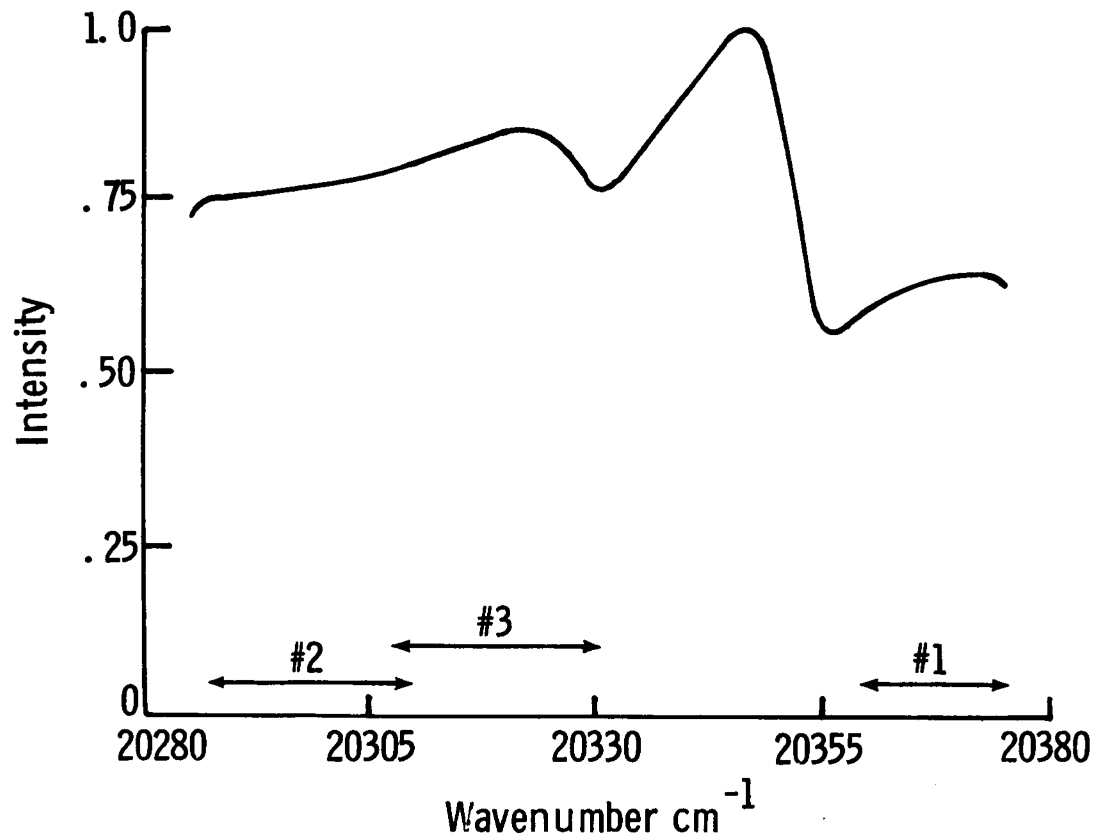


Fig. 7 Oxygen Q-branch spectrum at 3% concentration in volume for 1500K: 97% nitrogen

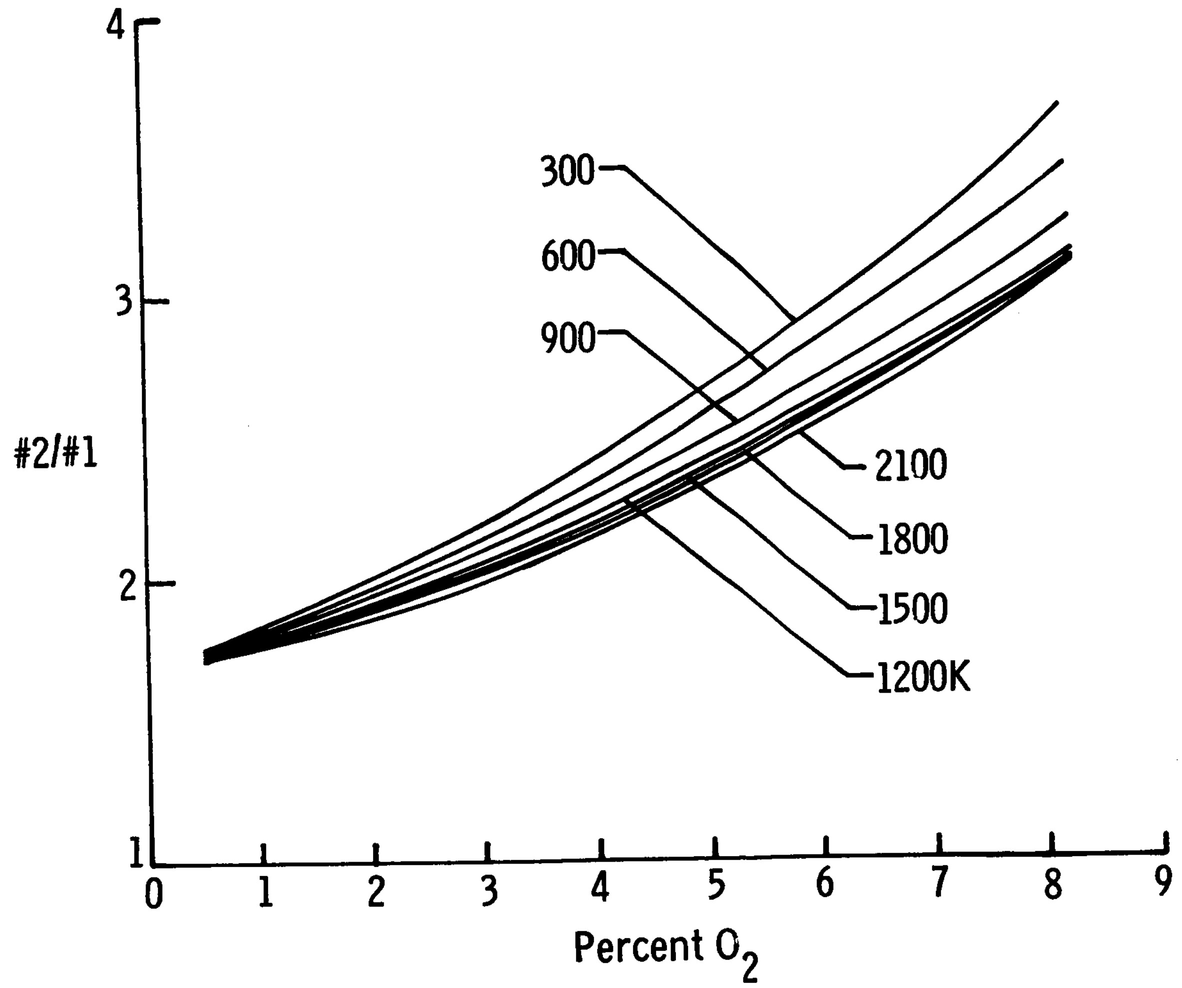


Fig. 8 The calculated intensity ratio of slit #2 to #1; #1 = 20360 ~ 20375cm-1, #2 = 20287 ~ 20312cm-1

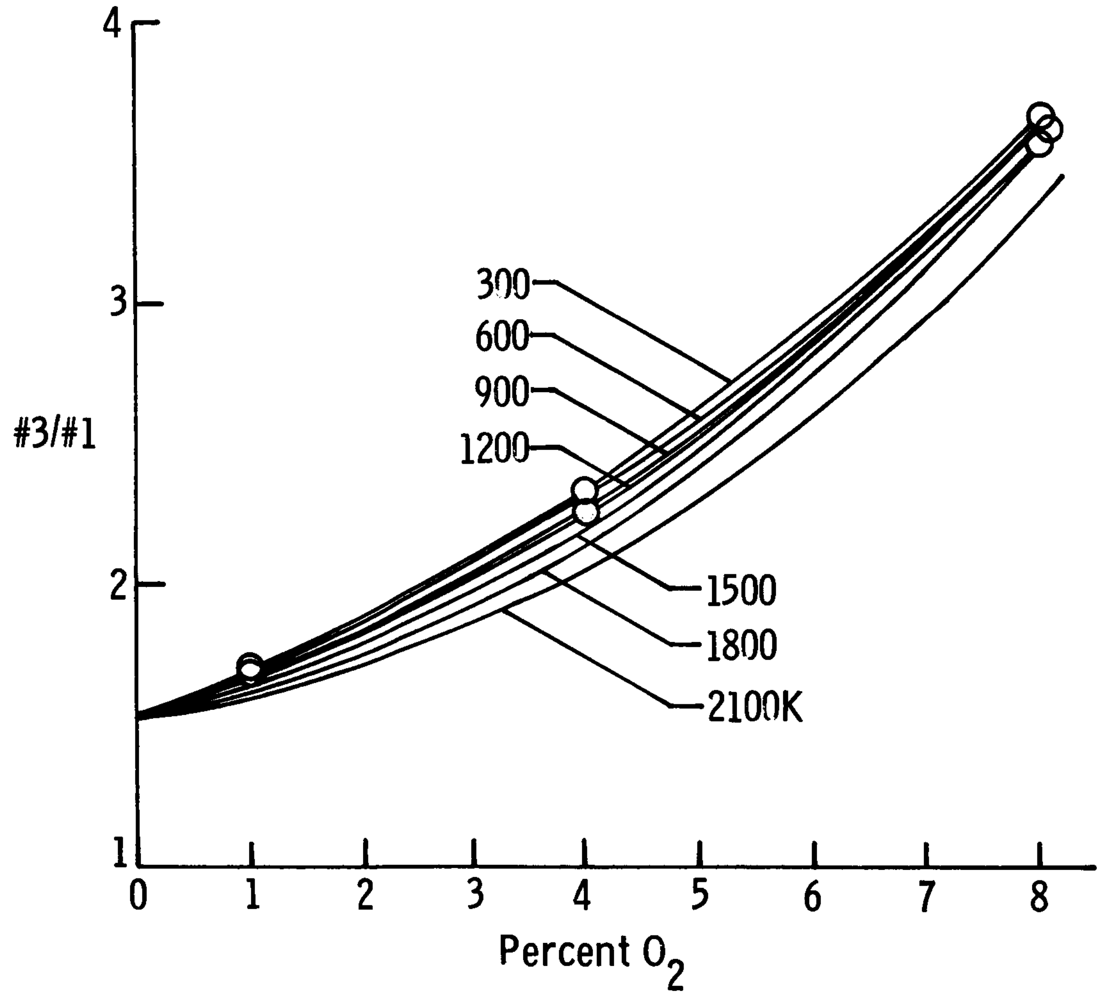


Fig. 9 The calculated and measured intensity ratio of slit #3 to #1;

#3 = 20307 ~ 20330cm-1

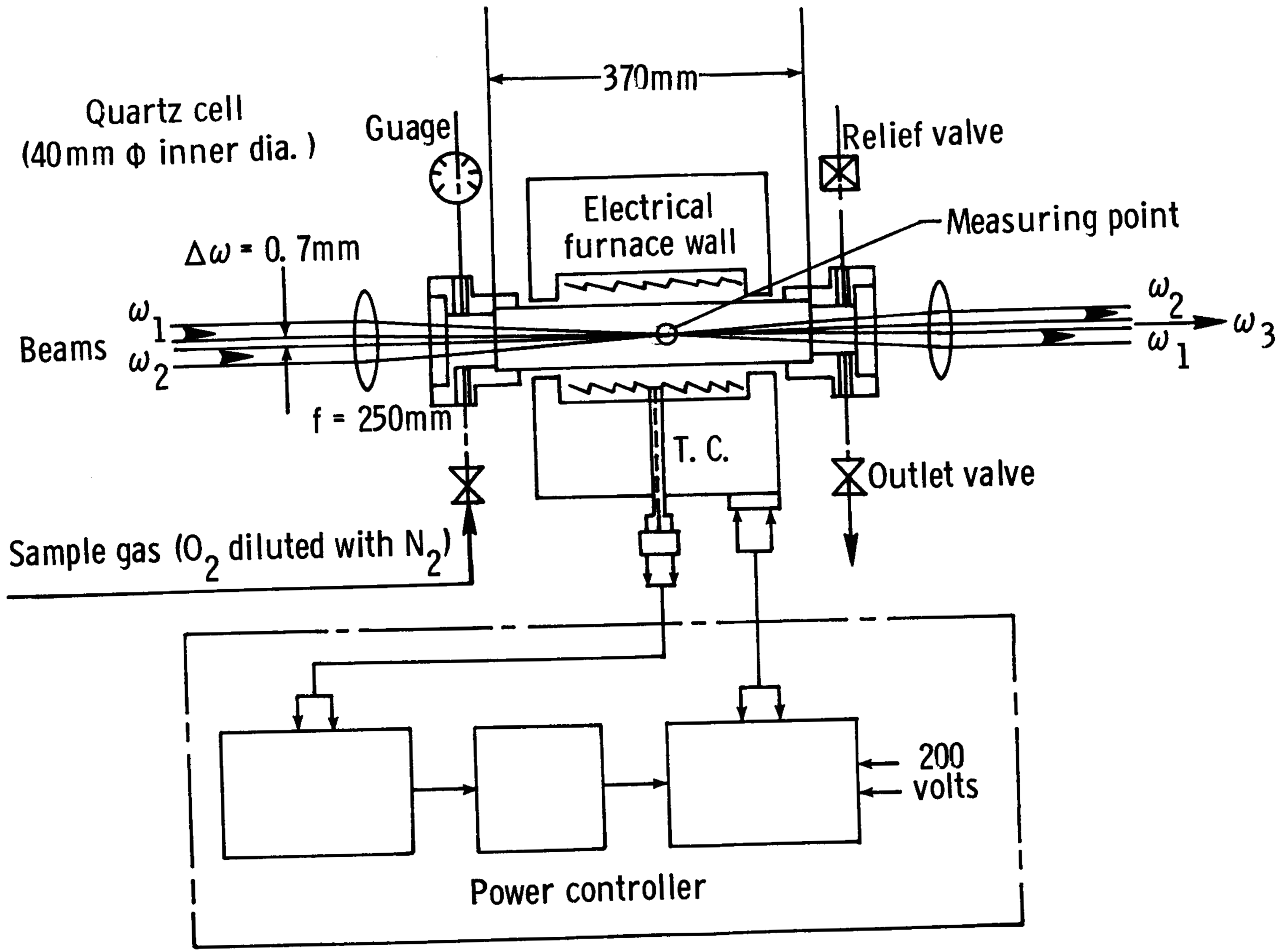


Fig. 10 Electric furnace and measurement setup for O_2 detection

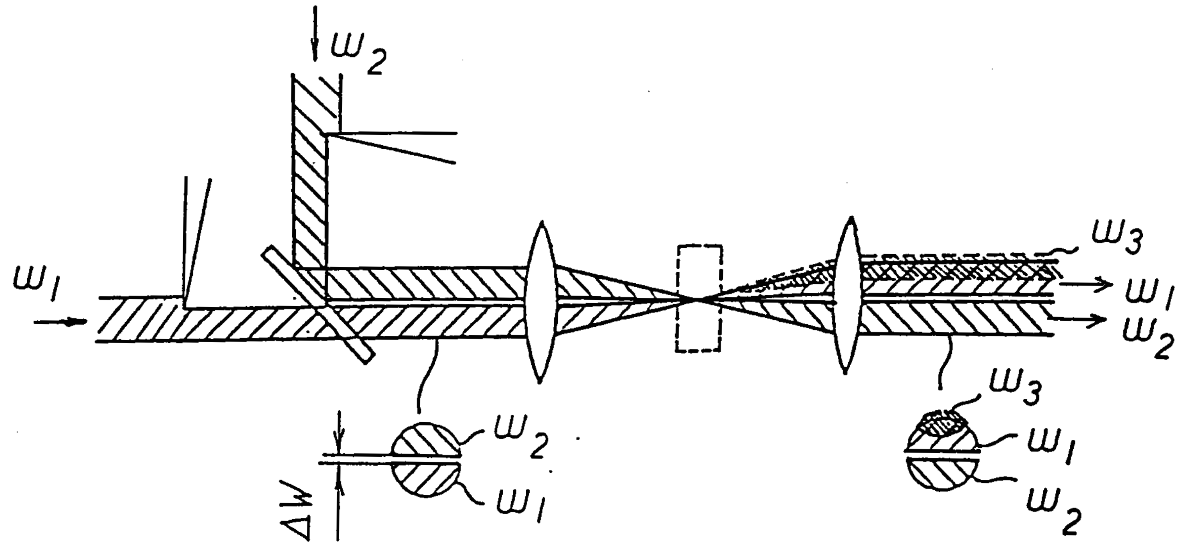


Fig. 11 MAC (Minim Angle Crossed-beam) alignment

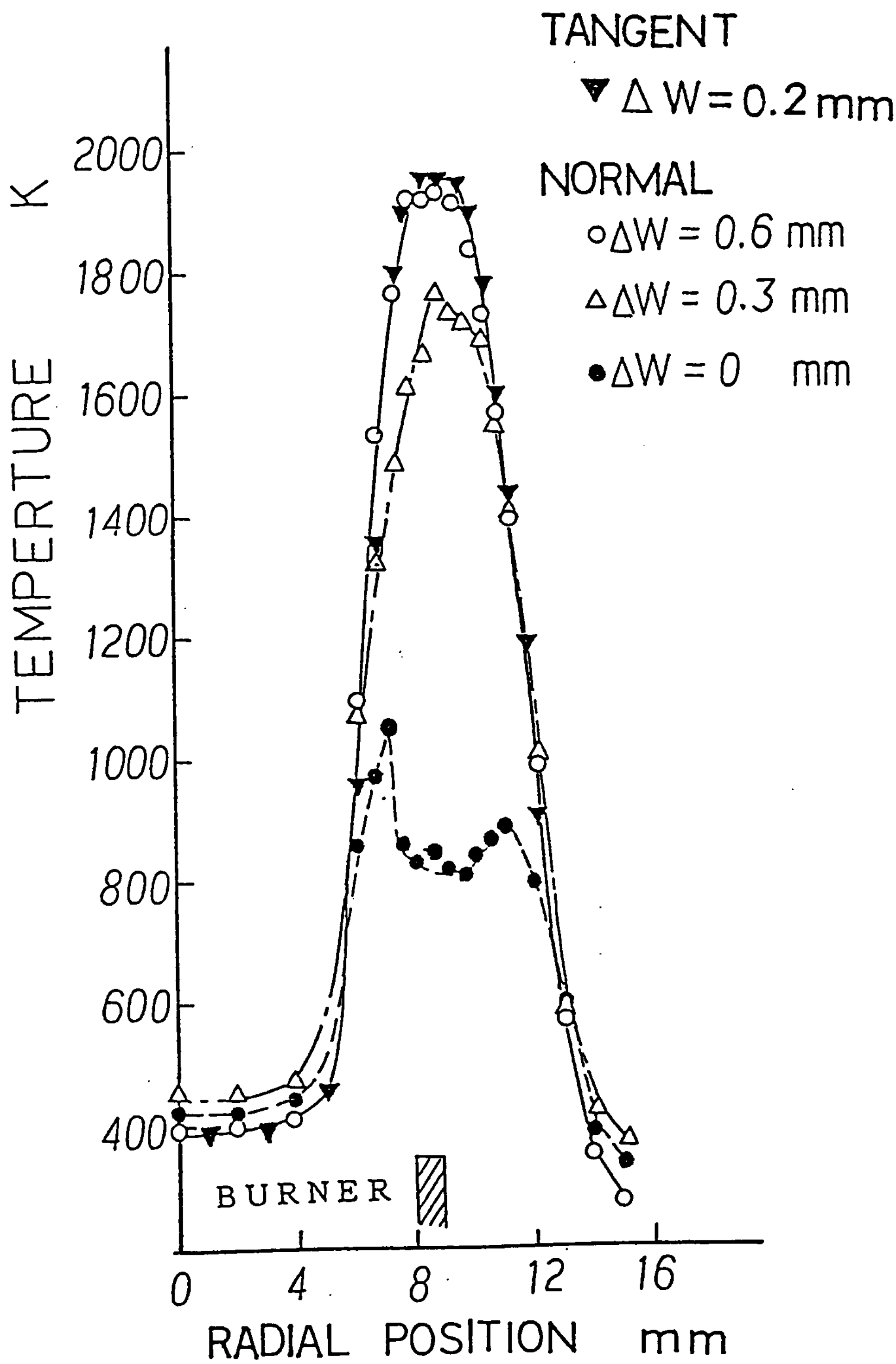


Fig. 12 Measured temperatures in Bunsen burner

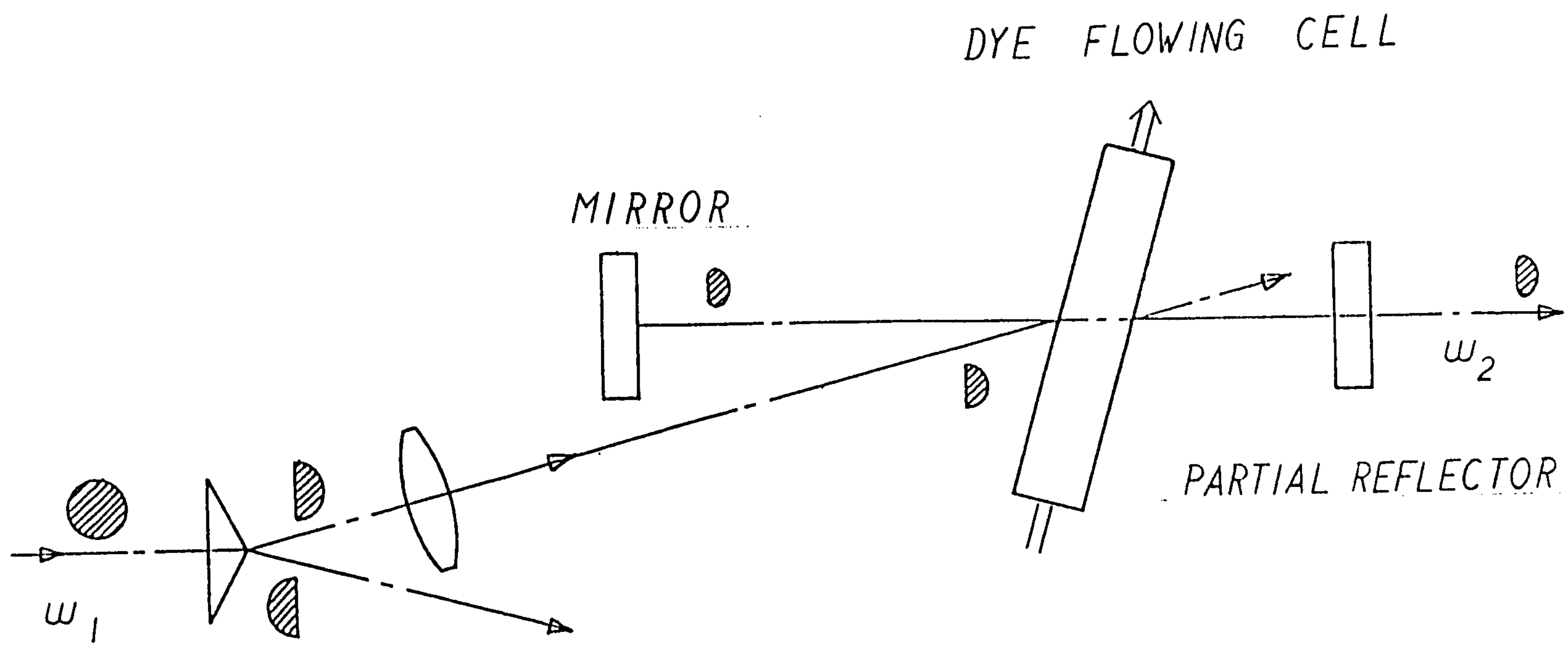


Fig. 13 Beam split by prism

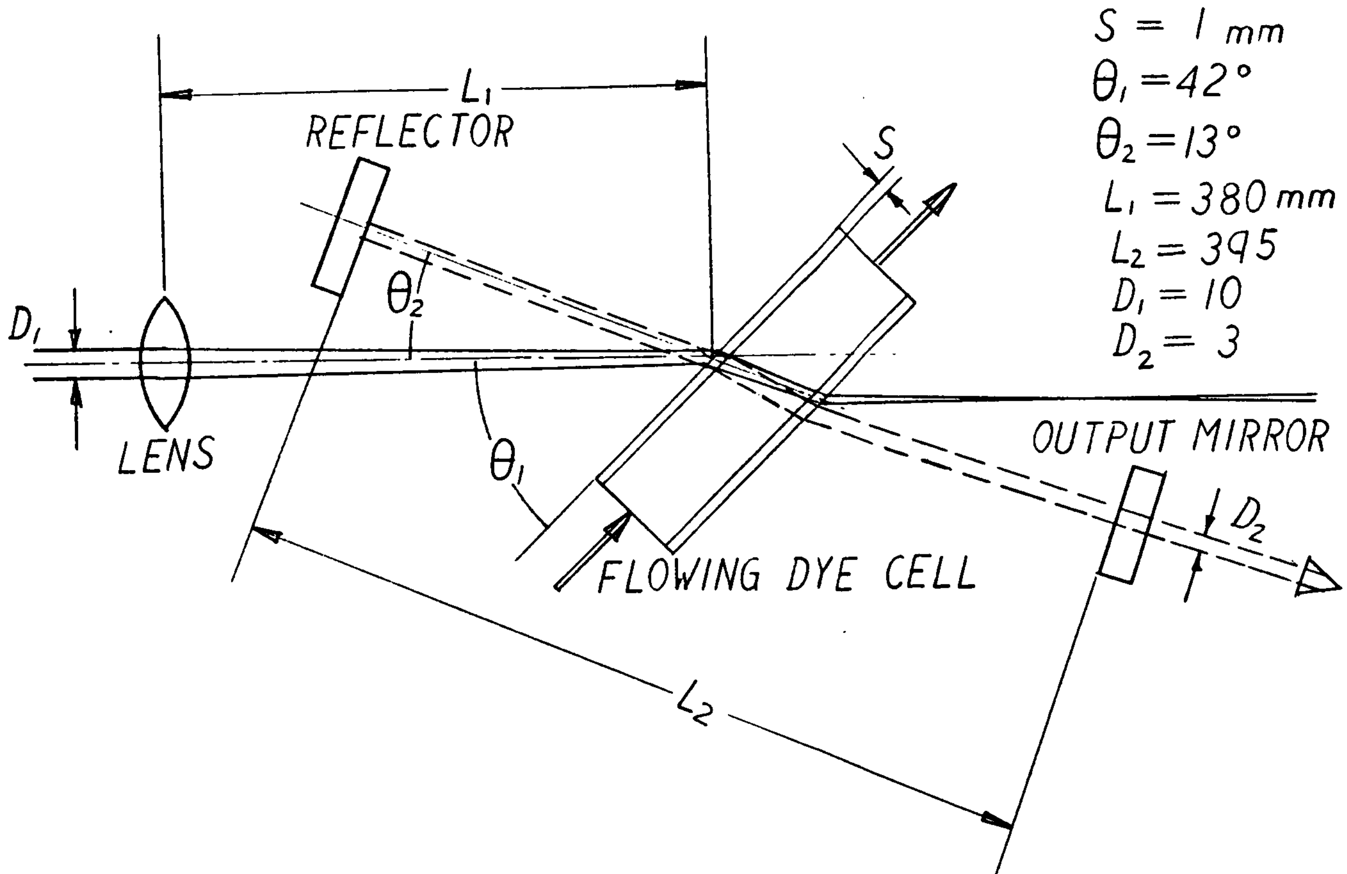


Fig. 14 Dye oscillator

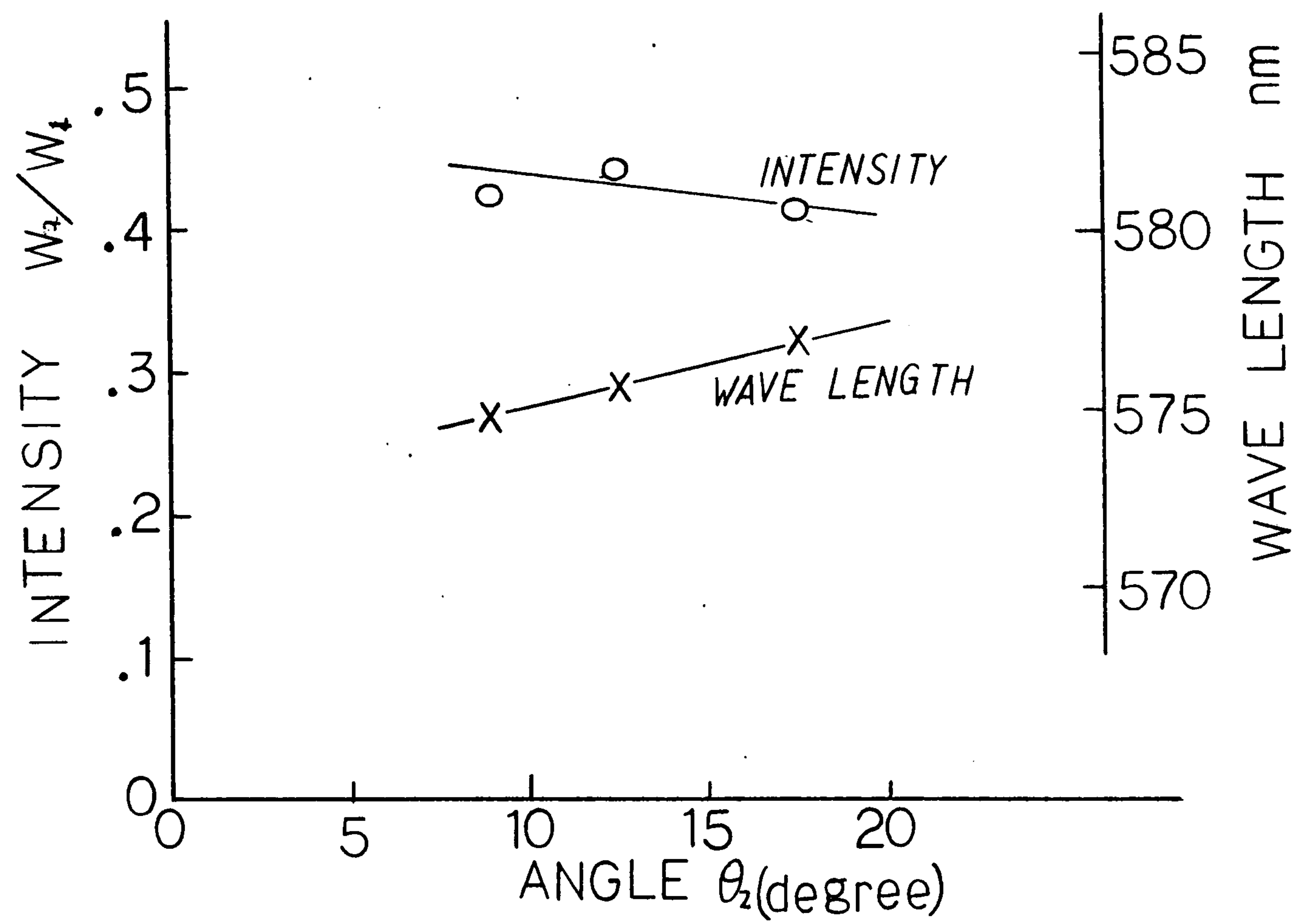


Fig. 15 Effect of angle between pump and Stokes beams on laser characteristics; Rhodamine 610

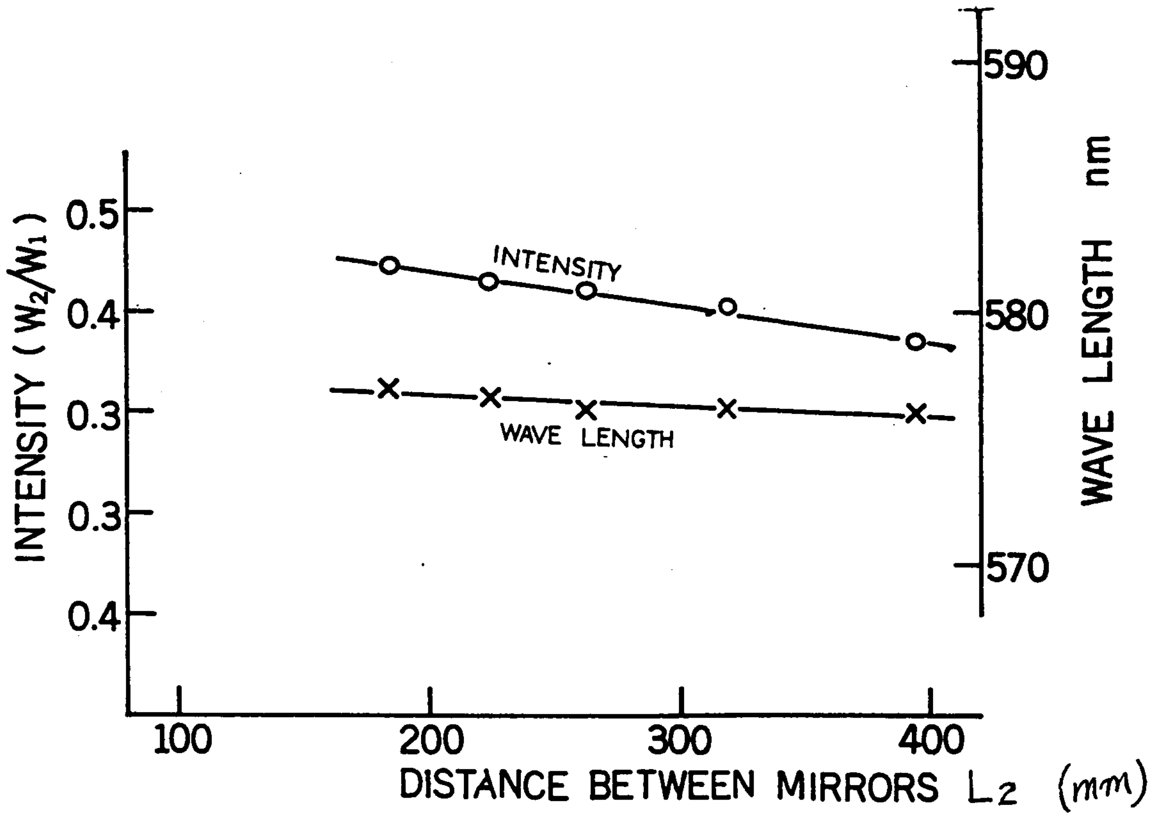


Fig. 16 Effect of resonant length

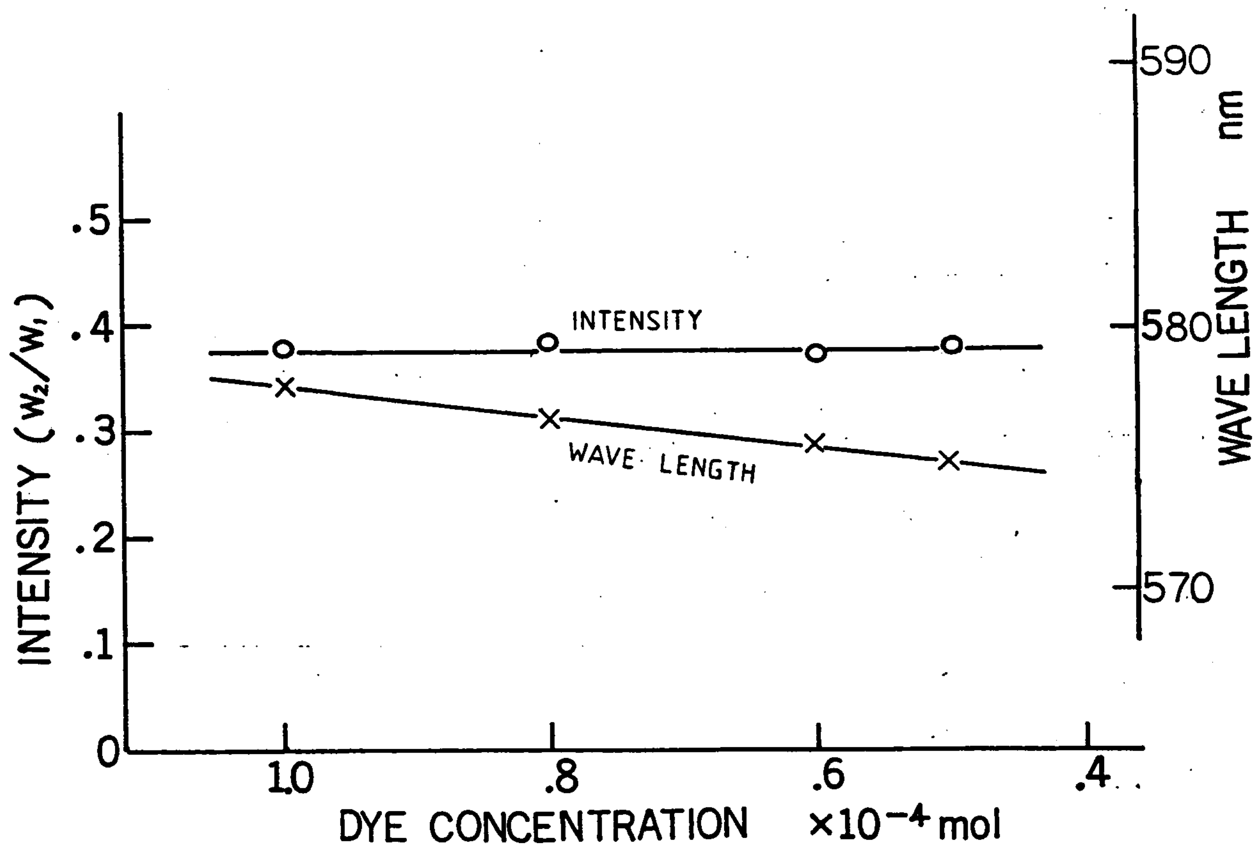


Fig. 17 Effect of dye concentration

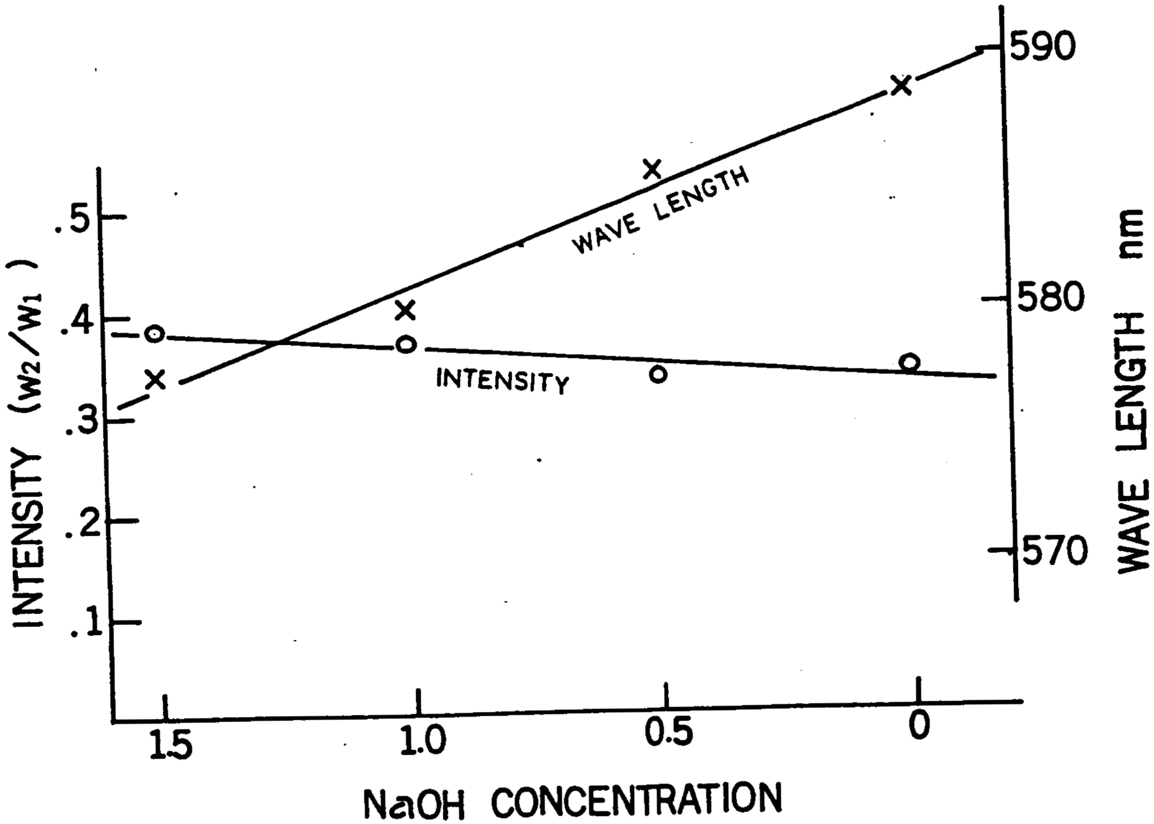


Fig. 18 Effect of NaOH addition

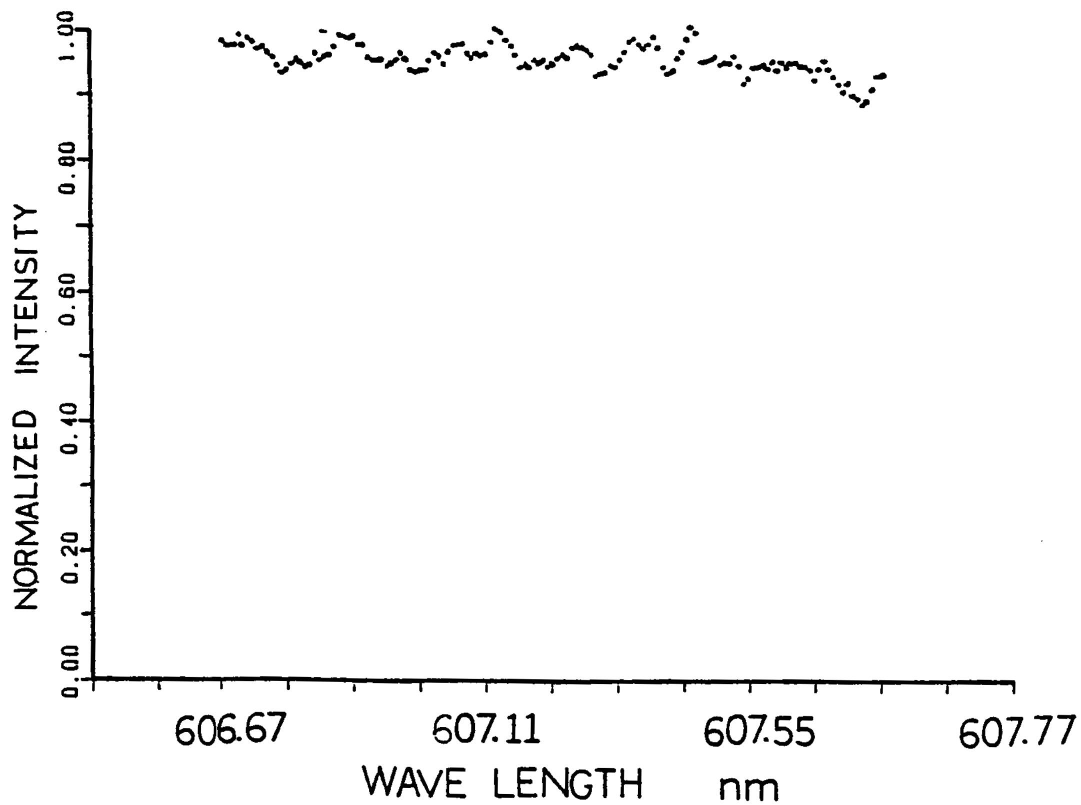


Fig. 19 Broadband profile of Stokes beam using Rhodamine 640

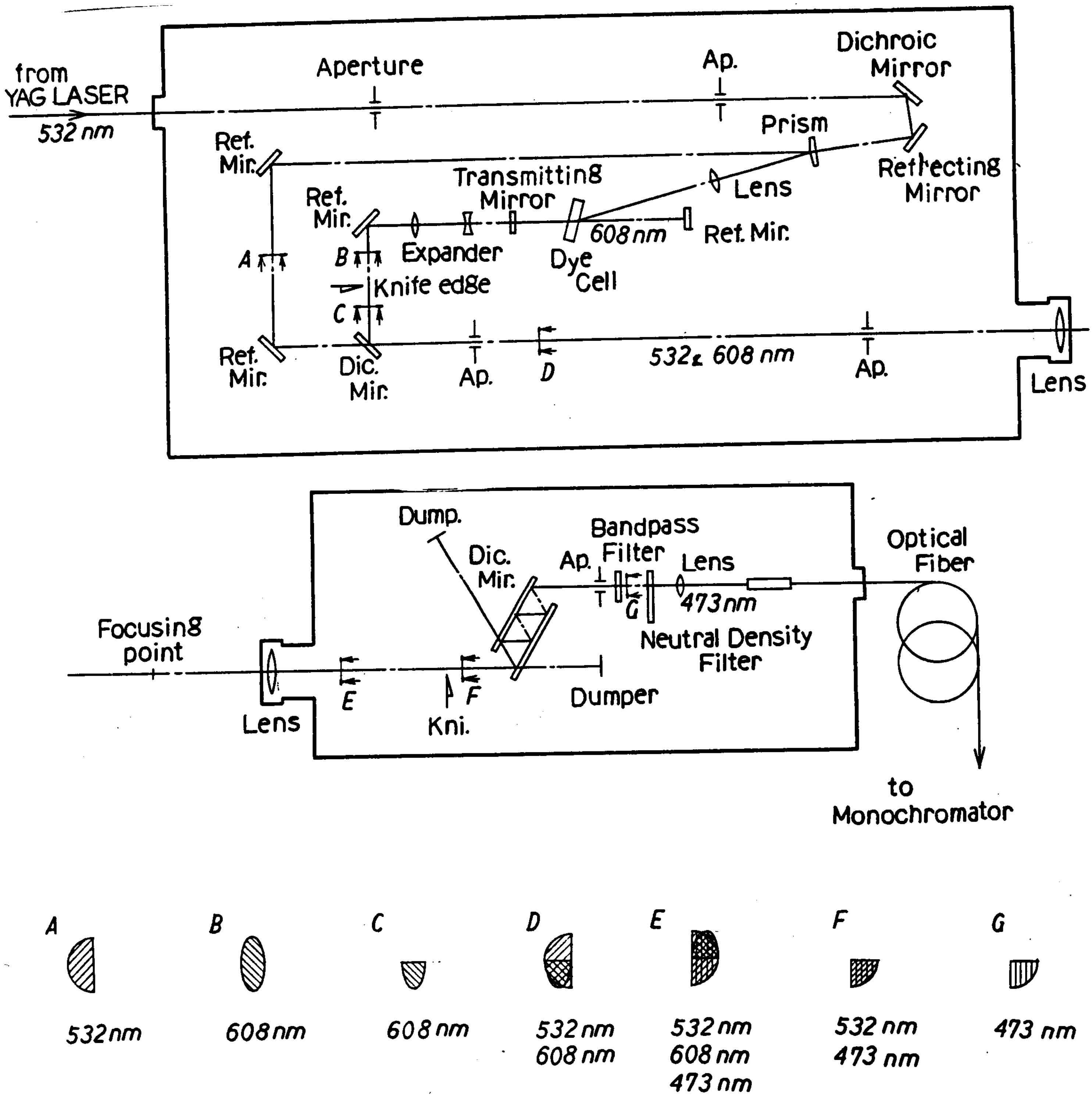


Fig. 20 Optical configuration for proposed simplified CARS measurement system

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15. Supplementary Notes

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