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Application of a two-color dye laser in CARS experiments for fast determination of temperatures

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In the specific case $M = \dot{N}$ and $N_{FP} = N_F$, which means that the dominant noise in the two instruments is the detector noise ($K \gg N$), we find the well-known Fellgett advantage $A = (N/8)^{1/2}$.

In general, for $M = N$ there is a limit ($N = 8$) below which a scanner is more efficient than the Fourier spectrometer. For $M < N$ the above limit is higher than 8 and depends on K , i.e., on the level of the background.

Figure 1 shows the behavior of A vs N for various K and M ; all the points which lie over the value $A = 1$ represent situations in which the Fourier interferometer is more efficient than a Fabry-Perot.

We must also note that the value of Eq. (4) can be improved using more complicated Fourier interferometers⁵ with $\gamma_F > 1$, which include optical configurations not possible with the Fabry-Perot. As an example, in astronomical observations, using detectors with a $DN \approx 10^{-15} \text{ W}/\sqrt{\text{Hz}} = \sqrt{K} BN$, in the millimetric window between 7 and 10 cm^{-1} , with 5 Torr of water vapor content, and with 1 cm^2 sr of throughput, we have a BLIP = $\sqrt{N} BN = 0.8 \times 10^{-14}$.

Using the definition of K we obtain

$$\sqrt{K} = \frac{DN}{BN} = \frac{DN}{\text{BLIP}} \sqrt{N}.$$

For $N = 10^4$ the Michelson is more efficient than a Fabry-Perot; in fact $K = 156$, and the actual advantage, computed from Eq. (4), is $A \approx 4.4$.

In other words the two instruments share the same SNR when $M = [8(K + N)]/(K - 1) = 504$; that is, when we are interested only in ~ 500 spectral elements.

We wish to thank B. Carli for useful discussions and suggestions.

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3. For further details see Ref. 2.
4. $\gamma = \alpha\alpha G_1 G_2$ defined in Ref. 2.
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Application of a two-color dye laser in CARS experiments for fast determination of temperatures

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The application of coherent anti-Stokes Raman scattering (CARS) as a measuring tool in combustion diagnostics is well documented (e.g., Ref. 1). In the first CARS measurements the wavelength of a dye laser at frequency ω_s was scanned through the wavelength region of the Stokes Raman band, determined by a fix-frequency laser at ω_p . The Raman shift of the studied molecule is then given by $\omega_p - \omega_s = \omega_R$. The CARS signal is generated through the third-order nonlinear susceptibility $\chi^{(3)}$ and observed at $\omega_{AS} = 2\omega_p - \omega_s$. These early CARS measurements were clearly limited to

stationary phenomena, e.g., laminar flames. In 1976 Roh *et al.*² showed that a complete CARS spectrum could be captured in one laser pulse by using a broadband ($\sim 150\text{-cm}^{-1}$) dye laser and a spectrograph followed by optical multichannel detection. A broadband laser profile can easily be obtained either by using the dye-laser grating in zeroth order or by replacing the grating with a totally reflecting mirror.

The CARS technique with the broadband approach has been used in various real-world applications, e.g., Refs. 3-7. In some of these experiments the aim was to measure probability distribution functions (pdf's) of temperature, which require at least a thousand single-shot CARS spectra to be statistically significant. The temperature from each single-shot measurement is normally evaluated by regression analysis of the experimental and theoretical generated CARS spectra. Clearly, with thousands of spectra this is a delicate task even for a very advanced computer.

The time-consuming analysis of the broadband CARS spectra was avoided by Eckbreth *et al.*⁸ by using different quick-fit methods. Then only parts of the broadband spectra were evaluated and compared to theory. In one such quick-fit method the temperature pdf was obtained by taking the ratio of the intensity of the fundamental band to that of the hot band of the studied N_2 molecule and comparing these ratios with those obtained from theoretical spectra.

In this Letter we report on an alternative quick-fit technique. Instead of using a broadband dye laser we have used a dye laser with only two wavelengths. The two-wavelength operation of the laser was achieved by using a wedge in the dye-laser cavity. With a proper inclination angle of the wedge, which covered half of the expanded laser beam in the cavity, it was possible to get a dye-laser beam with wavelengths centered at the fundamental band and at the hot band of the N_2 molecule, respectively. The approach of using a wedge in the dye-laser cavity to achieve lasing at two wavelengths was shown in 1975 by Schmidt.⁹ We have modified a commercial Quanta-Ray PDL-1 dye laser for our measurements. The arrangement is shown in Fig. 1. The pump beam at 532 nm is focused on the dye cell, and the dye-laser cavity beam is expanded by a prismatic beam expander to a sheet of $\sim 1 \times 25$ mm. The wedge, which is made of quartz and has an inclination angle of $\sim 0.8^\circ$, was then inserted into the beam as shown in the figure. Half of the beam is refracted in the wedge and meets the grating in a slightly different angle than the unrefracted light, thus producing a different wavelength.

The experimental setup for testing the two-color CARS technique was similar to the one described in Ref. 10 except for the wedge in the dye laser. A Quanta-Ray DCR-1A Nd:YAG laser, producing ~ 220 mJ at 532 nm in 6-nsec pulses, was used. Ten percent of this power was split off and served as

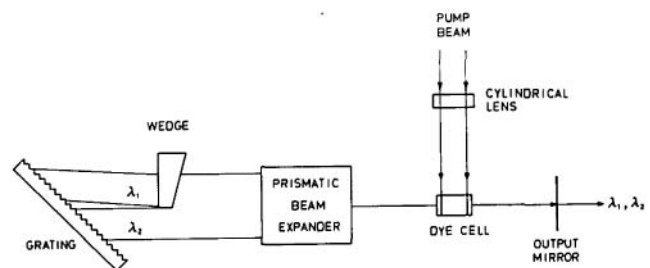


Fig. 1. Experimental approach for producing two wavelengths using a wedge.

a pump beam at ω_p in the CARS process, whereas the rest of the green light was pumping the dye laser producing a Stokes beam at ω_s of ~ 25 mJ around 607.3 nm. The laser beams were then aligned according to the BOXCARS approach,¹¹ permitting spatially resolved measurements, and focused in a premixed CH₄/air flame with a $f = 30$ -cm lens. The laser beams, two at ω_p , one at ω_s , and the CARS beam at ω_{AS} , were recollimated with a second $f = 30$ -cm lens. After the lens a dichroic mirror and an interference filter (HWHM = 10 nm), centered at the N₂ anti-Stokes wavelength at 473.3 nm, were used to isolate spectrally the CARS beam. The CARS beam was then directed to a 2-m Ebert spectrograph with a dispersion of ~ 1 Å/mm in fifth order and detected with a Tracor Northern diode-array detector TN-4IG, which was placed at the exit plane of the spectrograph. The detector array, which consists of 1024 diodes (2.5 mm \times 25 μ m), is placed behind an image-intensifier stage, and the system is gateable down to 0.5 μ sec for background suppression. To synchronize the laser pulse with the detector gating and readout, a special master trigger unit was constructed. The recorded CARS signals were stored on a Logic Data P80 CPM computer for subsequent data processing.

The two dye-laser beams were analyzed, and the wedge in the dye-laser cavity was adjusted by monitoring in real time with the diode-array detector. To check the dye-laser fluctuations from shot-to-shot a reference CARS setup was added to the experiment by splitting off laser light to a cell with argon at 7 atm. CARS beams were produced in the cell at two wavelengths through the nonresonant background of the third-order susceptibility $\chi_{NR}^{(3)}$. The signal and reference CARS beams were focused at the entrance plane of the spectrograph, where the slit was rotated 90° to allow all the beams to be dispersed and recorded beside each other on the detector array.

An experimental result is shown in Fig. 2, which is a photograph of the Tracor Northern display. The CARS signals corresponding to the fundamental and to the hot band of N₂ molecules in the flame are shown to the left in curve *a*, and the corresponding reference peaks are shown to the right in this curve, which is a single-shot registration. A CARS spectrum measured with the dye laser in a scanning mode is shown for comparison in curve *b* in the figure. This spectrum also served as reference, determining the resolution which is to be used for the computer-generated CARS spectra. It was also used as a wavelength scale for the dye-laser beams, since the ratio of the intensity of the fundamental band to that of the hot band is critically dependent on the wavelengths used. As pointed out by Eckbreth *et al.* in Ref 8, generating accurate reference spectra for normalization with a multimode pump laser is not trivial due to mode-hopping of the dye laser. The normalization procedure described here is a rough approximation.

Since no isothermal tube furnace was available, the premixed methane/air flame was used as a constant temperature object. These preliminary single-shot experiments revealed a mean temperature of 2075 K with a standard deviation of 200 K. The standard deviation of the temperature is partly due to the fact that the methane/air flame was not a stable source. Furthermore, it was realized that the diode-array detector suffers from severe saturation. Thus the CARS signals had to be attenuated to ~ 600 counts/shot; i.e., the detector shot noise was not negligible. This difficulty has been discussed previously by other groups in CARS experiments with a similar detector array.¹²

Clearly, the approach described above cannot be as accurate as a complete regression analysis, but it has certain distinct advantages. First, as stated, the computer time will be con-

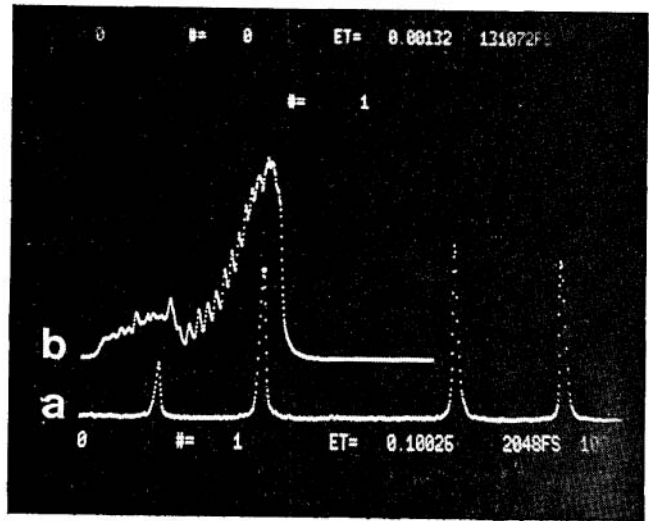


Fig. 2. Display of the optical multichannel analyzer: (a) CARS signals at two wavelengths produced in the flame (left) and the corresponding reference signals (right); (b) CARS spectrum measured with a scanning dye laser to determine the resolution and exact positions of the two wavelengths.

siderably lower. Second, the signal strength can be much higher than in a broadband measurement. In our case the signal was increased by a factor of ~ 20 , which will be advantageous in measurements in environments with high particle contents. We are currently planning to do measurements in a full scale coal furnace, where the soot attenuation may be severe. Furthermore, complex fuels may give large differences in $\chi_{NR}^{(3)}$. This can be overcome by applying a polarization technique which suppresses the $\chi_{NR}^{(3)}$ term.¹³ As the CARS signal then is reduced by an order of magnitude, the two-wavelength approach is favorable.

Spark diagnostics is another field where we want to apply this new technique. In this case a very high spatial resolution is required, and then the laser power is limited to avoid gas breakdown in the probe volume. The two-wavelength approach may then be useful, whereas the broadband CARS technique may give too low signal strengths.

Goss *et al.*⁴ and Eckbreth¹⁴ have demonstrated methods in which the problem with the large dynamic range in CARS measurements on turbulent combustion is partly overcome by splitting the beam of detection into several beams of decreasing magnitude. In the two-color CARS technique discussed here, their methods can be applied. The problem is minimized in our standard setup. The dynamic range in a CARS experiment is determined by the dynamic range of the fundamental band intensity and the one of the hot band studied. By decreasing the intensity of the dye-laser wavelength corresponding to the fundamental band and increasing the other one, the individual dynamic ranges are changed, and the resulting total dynamic range is decreased. The dye laser intensities are easily varied by simply changing the position of the wedge in the setup described.

The two-color CARS technique described here could, of course, be extended to, e.g., four colors, which in case of no background suppression would provide information about $\chi_{NR}^{(3)}$ also. Eckbreth and Hall¹⁵ have proposed such measurements of $\chi_{NR}^{(3)}$ using multiple-wavelength dye lasers, and measurements were demonstrated by Goss and Schreiber.¹⁶

Of course, the accuracy in CARS measurements using the two-color approach described in this paper is critically dependent on the frequency stability of the dye-laser components. We have made some investigations of this property by observing the spectrum on the Tracor display when using the spectrograph grating in the highest possible order. In these tests we concluded that the stability of the dye-laser wavelength on a shot-to-shot basis is better than 0.025 Å. However, on a longer time scale, the dye-laser wavelength may be changed due to temperature fluctuations of ~ 0.1 Å/C°.

Work is now under way in our laboratory to further evaluate and refine this technique. The applicability to measurements in large burners and spark diagnostics will be examined.

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Feature enhancement of film mammograms using fixed and adaptive neighborhoods: correction

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The 2-D Fourier transform magnitude spectra for the images in Figs. 4, 6, and 7 of this paper¹ were supposed to be given in Figs. 9(a)–(c) according to the text (p. 563), but they are missing. They are now given here numbered correctly as Figs. 11(a)–(c).

Note also that Figs. 9 and 10 on p. 563 are upside down.

Reference

1. R. Gordon and R. M. Rangayyan, "Feature Enhancement Using Fixed and Adaptive Neighborhoods," *Appl. Opt.* **23**, 560 (1984).

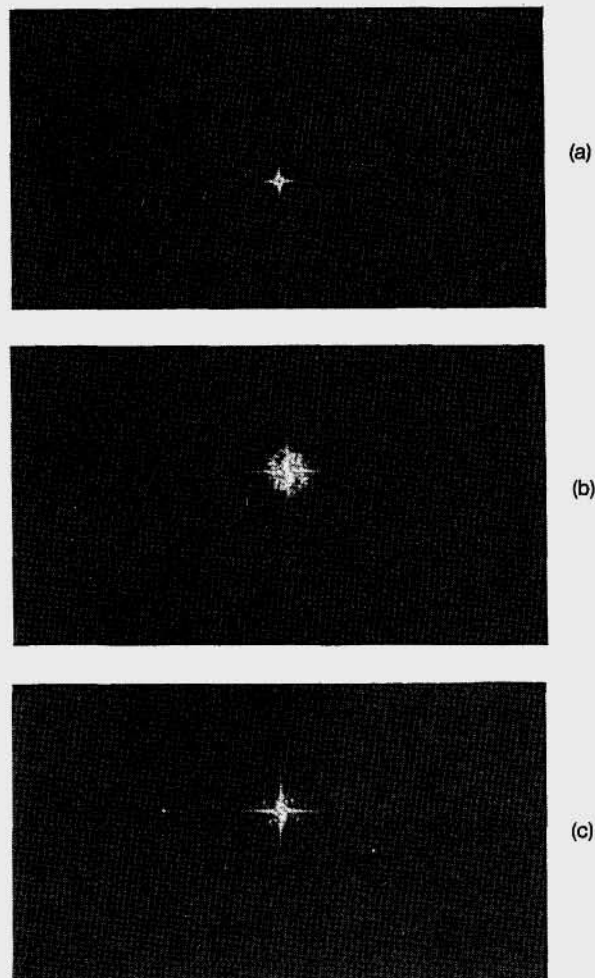


Fig. 11. (a)–(c) Log(1 + magnitude) spectra of images in Figs. 4, 6, and 7, respectively. The values were linearly transformed to the 0–255 range (integers). The values in the 0–63 range were stretched to the full range of display (0–255), with density clipping to 63 of those few values above 63. This was done to make the weak high frequency components visible.