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## CARS experiments in a full-scale (10 x 10 m) industrial coal furnace

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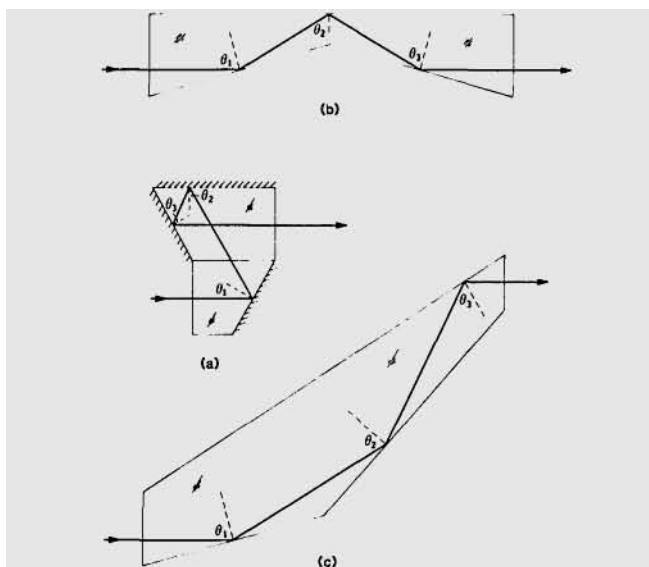


Fig. 2. Three-reflection prisms that, in combination with a single Porro prism, make a retroreflector. All satisfy  $\theta_1 + \theta_2 + \theta_3 = 90^\circ$ , where  $\theta_2$  in (b) and  $\theta_3$  in (c) are negative.

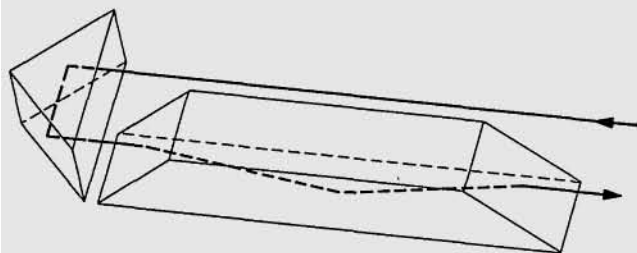


Fig. 3. Form involving only three reflections but requiring in addition two refractions.

gives reversion. It is not a practical system, however, since one of the reflections at  $45^\circ$  must be at a beam splitter to allow the light out.

But it is not necessary to restrict the angles of incidence to  $45^\circ$  and  $90^\circ$ . A suitable practical system can be made consisting of two reflections at  $45^\circ$  in a vertical plane, as in a single Porro prism, followed by three reflections in the horizontal plane. Figure 2 shows three possible arrangements of these three reflections. The three angles of incidence all satisfy the relation

$$\theta_1 + \theta_2 + \theta_3 = 90^\circ, \quad (6)$$

where the angle is positive if it deviates the light in a positive sense (anticlockwise). All the cases have been drawn as prisms, but mirror systems are equally suitable.

These systems all involve five reflections, and any retroreflector must have an odd number. The minimum possible number is three, as in the cube corner. But when a ray is incident at a cube corner in a plane containing the normals to two of the surfaces, it is parallel to the third surface and so never meets it. The arrangements of Fig. 2 replace this third mirror by mirrors that the ray will meet.

The ray can also be made to meet the third mirror of a cube corner by refraction. The combination, shown in Fig. 3, of a single Porro prism and a Dove prism is also a retroreflector

that preserves horizontal and vertical linear polarizations. The two refractions in Fig. 3 replace two of the reflections in Fig. 2.

If the system is to satisfy the stricter condition that it have no effect on any polarization, the phase difference produced by the two vertical reflections at  $45^\circ$  should match that due to the three horizontal reflections. For front-surface mirrors or aluminized prisms such matching depends on the conditions under which the surfaces are coated as well as on the materials used. It should be possible to correct systems of the type in Fig. 2(a). But a surer correction can be obtained with prism systems that rely entirely on total internal reflection, for which the phase changes are much more reproducible. For this correction the less compact forms [Figs. 2(b) and 2(c)] must be used. If the glass has a refractive index of 1.54, two reflections at  $45^\circ$  are corrected by three reflections at  $75^\circ$ ,  $75^\circ$ , and  $-60^\circ$ . The system will then transmit any polarization with its ellipticity unchanged, for example, circularly polarized light remains circular. But, because of the reversion, right-handed is changed to left-handed as by a plane mirror. Hence a quarterwave plate in front of such a retroreflector will change horizontal linear polarization to vertical and so allow the forward and returned beams to be separated by their polarizations. This technique is important for plane mirrors but less so for retroreflectors where the beams are also separated spatially.

Unlike the cube corner, for which a ray aimed at its apex returns along the same path, all the systems described in this paper return every ray with a lateral displacement. The center of the system, from which the incident and reflected ray are equally distant on opposite sides, lies outside the in and out beams. Its position can be adjusted by a change of size of the component prisms (or a change of mirror separations).

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## CARS experiments in a full-scale ( $10 \times 10$ m) industrial coal furnace

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Coherent anti-Stokes Raman scattering (CARS) has, since the basic theory was outlined in the Sixties, proved to be a powerful technique for probing molecular properties. Its application to combustion diagnostics was proposed by Taran<sup>1</sup> more than ten years ago. Since the early work on  $H_2$  molecules in a stable well-defined  $H_2$ /air flame, the technique has been developed for single-pulse measurements<sup>2</sup> and also for high spatial resolution measurements by the introduction of the BOXCAR technique.<sup>3</sup> With these improvements the CARS technique has been used in several real-world combustion applications; see, e.g., Refs. 4–7.

In this Letter we report on what we believe to be the first CARS measurements in a full-scale ( $10 \times 10$  m) coal fired boiler. Reports have previously been made on a semi-industrial oil furnace.<sup>8</sup>

Before going into the experiment in detail, a brief account of the basic CARS concept will be given. For a more compre-

hensive review we refer the reader to Refs. 9–11. In the normal CARS approach two laser beams at frequencies  $\omega_p$  and  $\omega_s$  are crossed and focused in the combustion zone. When the frequency difference  $\omega_p - \omega_s$  is equal to a molecular vibrational (or rotational) energy splitting which is Raman active, a CARS beam is created through the interaction of four photons through the third-order nonlinear susceptibility  $\chi^{(3)}$ . In addition to the frequency requirement of the laser beams for CARS generation, the phase-matching condition has to be fulfilled. The easiest way to do this in gases, which have almost zero dispersion, is to use a collinear approach; i.e., the two laser beams are superimposed and have identical pathways. The drawback with this approach is that the spatial resolution is low. As has already been mentioned, this problem was solved by Eckbreth through the introduction of the BOXCARS concept,<sup>3</sup> where the laser beams are split up and CARS generation only occurs from the region where the beams cross.

The CARS signal at  $\omega_{AS} = 2\omega_p - \omega_s$  is generated as a new laser beam from the focal region, and by using a simple aperture discrimination against even a hostile environment can be achieved.

The CARS experiment reported in this letter was carried out at a coal fired boiler which provides heat to a district in southern Sweden. The boiler, which was constructed about thirty years ago, has a total capacity of more than 150 MW heat at full load. To be well prepared for CARS experiments over the long distance involved with this furnace (10 m), preliminary experiments were performed in the laboratory, where signal strength and spatial resolution were investigated for collinear CARS and USED CARS and for different beam diameters and focusing conditions. These tests revealed that the choice of lens to be used in the beam expander and the position where the laser beams hit the front lens were critical; e.g., with a small concentric lens position in the beam profile, astigmatism was introduced which lowered the CARS efficiency dramatically. During the experiments it was found that the best choice was to use collinear CARS with the laser beam expanded by a factor of 3 to a beam diameter of  $\sim 3$  cm by using one negative and one positive lens,  $f = -7$  cm and  $f = 20$  cm, respectively, in the beam expander. The laser beams were then focused with an inexpensive spectacle lens,  $f = 5$  m.

With the laboratory experiences in mind a movable CARS table was built, as shown in Fig. 1. A Quanta-Ray DRC-1A Nd:YAG laser was used as a pump laser producing  $\sim 250$  mJ/pulse at 10 Hz and 532 nm. Fifty percent of this green beam was split off and served as a pump beam at  $\omega_p$  in the CARS process, while the other 50% was used to pump a Quanta-Ray PDL-1 dye laser producing the Stokes beam at  $\omega_s$ . For probing  $N_2$  molecules a  $\omega_s$  value corresponding to 607.3 nm was used, and in our setup the laser power at this wavelength with rhodamine 640 dye was  $\sim 25$  mJ. The two beams at  $\omega_p$  and  $\omega_s$  were combined on a dichroic mirror, expanded, and focused with the beam expander and lens described above. The  $N_2$  CARS signal at 473 nm generated from the focus was isolated with a dichroic mirror, transmitting blue and reflecting green and red radiation, followed by a set of Corion low-pass filters. As detector either a PARC OMA2 optical multichannel analyzer or a PMT EMI 9558 was used at the exit plane of a home-built spectrograph,  $f/10$ , 2 Å/mm. During the laboratory experiments it was found that 90% of the CARS signal was generated from  $<50$  cm.

The coal furnace where the CARS experiments were carried out was  $10 \times 10 \times 30$  m and consisted of  $3 \times 3$  individual burners. The coal is injected as a fine powder, and the consumption is  $\sim 500$ – $600$  tons of coal per day. The furnace looks completely opaque, and it was barely possible to look

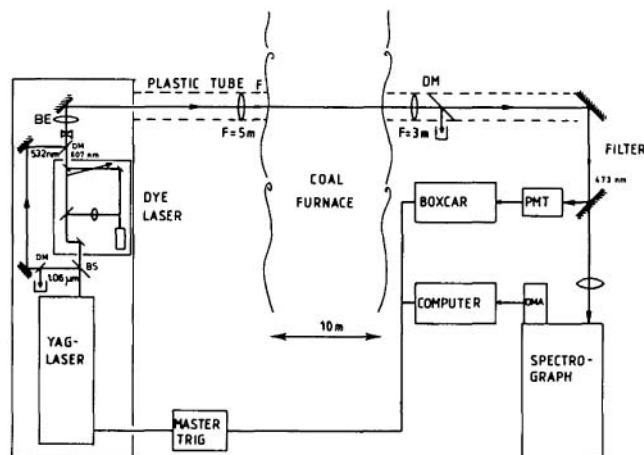


Fig. 1. Experimental CARS setup: BS, beam splitter; DM, dichroic mirror; BE, beam expander; F, optical filter.

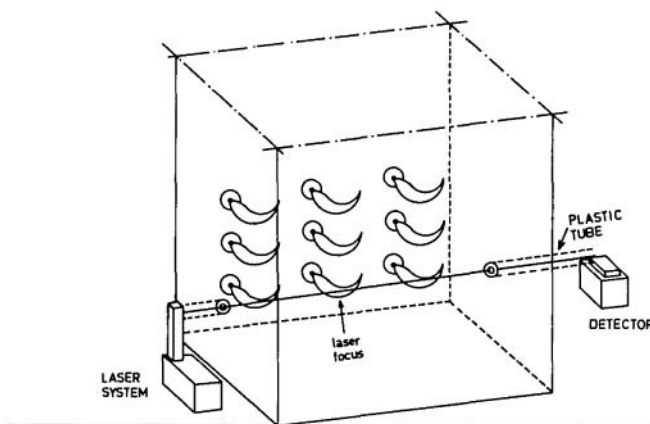


Fig. 2. Coal furnace and the laser/detection equipment.

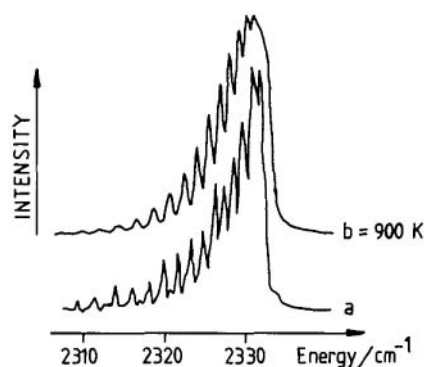


Fig. 3. a, Scanned CARS spectrum from the coal furnace; b, computer-generated CARS spectrum at 900 K. The resolution is  $1 \text{ cm}^{-1}$ .

Table I. Standard Deviation in Pulse-to-Pulse Fluctuations for the Green, Red and CARS Beam

|                    | 532 nm<br>(%) | 607 nm<br>(%) | CARS<br>(%) |
|--------------------|---------------|---------------|-------------|
| Before the furnace | 12            | 19            |             |
| After the furnace  | 23            | 29            | 56          |

into the furnace with the naked eye because of the intense glow of the coal particles.

In Fig. 2 the experimental CARS setup and the coal furnace with the nine burners are shown. The CARS setup was placed close to the furnace, and the laser beams were directed through a hole in the furnace wall ( $\phi \approx 10$  cm), which gave optical access to the furnace. Fortunately, the furnace had two holes which gave a straight line of vision through the furnace, which is necessary in CARS experiments. To protect the optics from dust and also for safety reasons all laser paths, before and after the furnace, were sealed with plastic tubes. To eliminate the risk of CARS generation from cold air outside the furnace, a GG 495 Shott long-pass filter was placed just before the laser beams entered the furnace. Fortunately, at the beginning of the experimental run the power plant was out of order, which made it possible to carry out the beam alignment more simply. This meant that when the furnace was running the laser beams were aligned so that both the green and the red beam could be seen at the exit hole of the furnace. The beam transmission through the furnace was measured and found to be  $\sim 5\%$ , and the laser beam appeared as a diffuse dancing spot with a sort of speckled pattern changing from shot to shot.

The first CARS experiments were carried out with a broadband dye laser and multichannel detection. However, it was not possible to detect any CARS signal using this technique. Instead a narrowband dye laser approach was tried, where the laser power at 607 nm with a bandwidth of  $\sim 0.4$  cm $^{-1}$  and measured just before the furnace entrance hole was  $\sim 6$  mJ, while the power of the 532-nm beam here was 40 mJ. (The relatively low laser power measured here, compared with those stated in the laboratory experiments, was mainly due to malfunctioning of the laser. However, losses in prisms used for directing the laser beams to the position of the furnace hole also contributed.) Now a CARS signal was clearly detected on the OMA console. To obtain a scanned CARS spectrum a photomultiplier was used as a detector, and a PARC model 4402 signal processor with a model 4422 gated integrator with a time window of 20 nsec was used. In Fig. 3 a CARS spectrum  $a$  from  $N_2$  molecules in the furnace is shown illustrating the fundamental band  $v = 0 \rightarrow v = 1$ . For comparison a computer generated CARS spectrum at 900 K is shown in Fig. 3b. The peak signal intensity in the CARS beam was  $\sim 50$  mV/pulse with the actual high voltage on the photomultiplier tube, which approximately correspond to on the order of 100 detected photons/pulse.

The computer generated CARS curve in Fig. 3 is not achieved by a fitting procedure. It is obvious that the conditions in the furnace change very rapidly, so that during the scan time the temperature also changes. This phenomenon makes the CARS curve look cooler, since a cool contribution is weighted stronger than a warm one.

The time constant when recording the spectrum  $a$  shown in Fig. 3 was 1 sec, and the recording was completed in 2 min. It was found that the intensity of the CARS signal exhibited large pulse-to-pulse fluctuations. To illustrate this,  $10^4$  single shots were recorded by using the time histogram function on the BOXCAR integrator. Similar distributions for the green and red laser beam were measured before and after the furnace, and the results are summarized in Table I.

As can be seen in Table I the CARS beam intensity exhibits large pulse-to-pulse fluctuations. However, these fluctuations are mainly due to those in the laser beams. Ideally, following the square and linear dependence of the green and red laser beam on the CARS process, the numbers stated above would give a fluctuation in the CARS beam of 44%.

The surprisingly large fluctuations in the two laser beams before the furnace were mainly due to a leaking Q-switch in the Nd:YAG laser cavity. A burnt dye-amplifier cell also contributed to the bad dye laser performance. Laboratory measurements in later experiments gave a pulse-to-pulse fluctuation at 607 nm of 6%.<sup>12</sup>

This Letter shows that it is clearly possible to make CARS measurements even in the aggressive and heavy particle-laden environment that a full-scale coal furnace constitutes. Before doing the experiments several potential problems were recognized, e.g., vibrations in the ground, problems with optics and electronics due to the dusty environment, beam attenuation in the furnace, and beam steering effects. None of these effects was experienced as being too severe. The dusty environment was protected by using plastic sheets over all electronics and optics including the laser system. It seems that the considerable beam attenuation is the most difficult problem to handle. The conclusion that the beam steering effect was not experienced as too severe is based on comparisons with the numbers in Table I and those measured in the laboratory under well-controlled conditions.

As has been mentioned, the CARS signal was too low to be detected using a broadband dye laser. We think that the signal strength can be increased so that single-shot measurements can be performed by: (a) Increasing the laser power for both the green and red beam. A practical limit for the highest laser power that can be used is when laser-induced breakdown occurs at the focus. For these large particles ( $\sim 100$   $\mu$ m) the threshold for laser-induced breakdown is decreased by  $\sim 2$  orders of magnitude compared with clean air.<sup>13</sup> (b) Using lenses with longer focal lengths in the beam-expander and also easier adjustment possibilities for their positions, this in order not to introduce astigmatism. (c) Using a focusing lens with high optical quality.

In the case of a too low signal for broadband CARS operation, an alternative could be to use a two-color dye laser as demonstrated in Ref. 14. Here a wedge is placed in the dye-laser cavity, refracting half of the expanded dye-laser beam, thus producing an additional wavelength. Then one wavelength is tuned to a rotational transition in the fundamental band,  $v = 0 \rightarrow v = 1$ , while the other wavelength is tuned to the hot band,  $v = 1 \rightarrow v = 2$ . This approach would yield a CARS signal intensity of the same order as shown in Fig. 3, thus making single-shot temperature and probability distribution function (pdf) measurements possible.

The present letter is a result of a project aimed at investigating the applicability of CARS in industrial burners. We have carried out experiments in several large-scale burners (oil, heavy oil and coal). The results, and further experimental details from this project will be given in a future publication (15).

The authors gratefully acknowledge the help of the staff at Öresundsverket in Malmö and also B. Edwall and H. Karlsson from Sydkraft AB. We are also grateful to H. Edner for assistance in using the PARC boxcar equipment. The general interest from K. Fredriksson and S. Svanberg is also appreciated. For evaluation of our CARS spectra, a computer program was kindly put to our disposal by R. J. Hall, UTRC.

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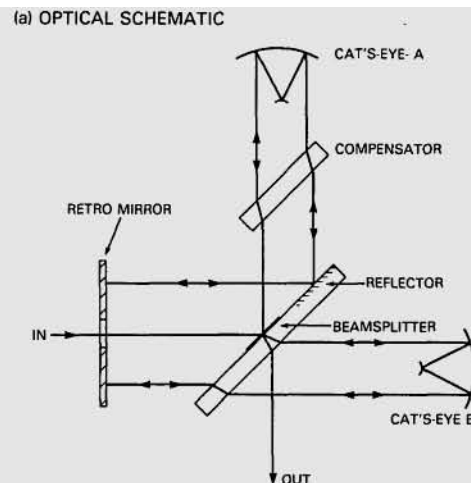
### Michelson interferometer with separated inputs and outputs, double pass, and compensation

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This Letter proposes a new configuration for a Michelson interferometer spectrometer having insensitivity to tilts or displacements of the parts, separated inputs and outputs, and double passing for higher resolution. It is a modification of the patented design<sup>1-5</sup> shown in Fig. 1, which was chosen for the atmospheric trace molecule spectroscopy (ATMOS) project and built at the Honeywell Electro-Optics Center for the Jet Propulsion Laboratory. The great advantage of compensated designs is a relaxation of mechanical tolerances, which is especially beneficial for instruments in hostile environments. The ATMOS, for example, must work reliably after the vibration of launch on the Space Shuttle. It is also beneficial when accurate instrument line profiles are required, for example, in studying unresolved molecular lines.

The ATMOS instrument operates as shown in Fig. 1. First, the incident ray splits at the beam splitter and both parts travel to normal cat's eye retroreflectors. However, the cat's eye portions used are not symmetrical, so the re-



(b) PATTERN AT BEAMSPLITTER SUBSTRATE

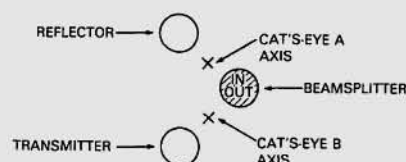
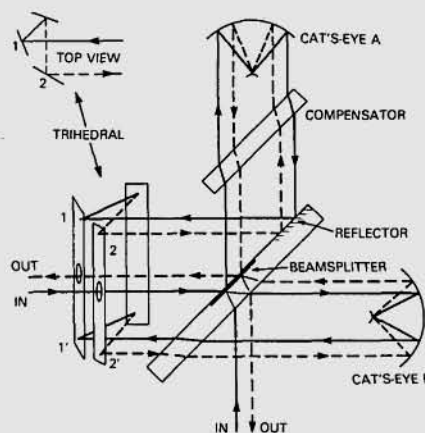


Fig. 1. Single input/output compensated Fourier transform spectrometer showing (a) optical schematic, and (b) arrangement of beams on the beam splitter as seen from the input.

(a) OPTICAL SCHEMATIC



(b) PATTERN AT BEAMSPLITTER SUBSTRATE

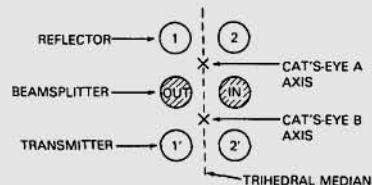


Fig. 2. Dual input/output compensated Fourier transform spectrometer showing (a) optical schematic, and (b) arrangement of beams on the beam splitter as seen from the inputs. The beam paths in (a) have been offset for clarity and actually have the pattern shown in (b). Cat's-eye A has 1, 2, OUT, and IN in its four quadrants, whereas cat's-eye B has OUT, IN, 1' and 2' in its quadrants.