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RESONANT CARS DETECTION OF HYDROXYL RADICALS(U) UNITED
TECHNOLOGIES RESEARCH CENTER EAST HARTFORD CT
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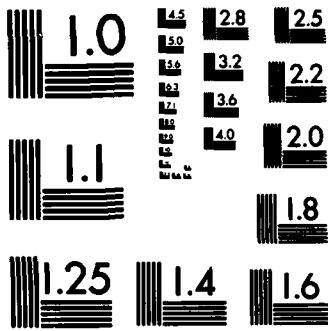


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been achieved for both systems. Moreover, an automatic tracking frequency doubling device which maintains constant UV power output as the fundamental frequency is tuned, has been installed on one of the dye lasers. A fine-scale, multielement diffusion burner using either H₂/O₂ or CH₄/O₂ has been tested for stability and uniformity. OH emission spectra have been observed for the 0-0 and 1-0 bands of the 2Π_{3/2} - 2Σ_{1/2} system. The search for resonant CARS in the OH radical is underway.

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East Hartford, Connecticut 06108

R82-955655

RESONANT CARS DETECTION OF HYDROXYL RADICALS

Annual Report

Contract No. F49620-81-C-0063

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Annual Report on Progress for Resonant CARS
Detection of the Hydroxyl Radical

Coherent anti-Stokes Raman Spectroscopy (CARS) is a proven diagnostic technique for both research scale and large size practical combustion devices. CARS offers a remote, non-perturbing method for accurately measuring temperature and species concentration with excellent spatial and temporal resolution. The numerous applications of CARS to highly varied combustion measurements has been well-documented and will not be listed here. A major limitation of CARS is the restriction to measurement of major species, typically to concentrations greater than ~0.5%.

The objective of this research program is to determine if CARS can be extended to minority species, such as radicals, by means of electronically enhanced resonant CARS. Electronically enhanced resonant CARS, or simply, resonant CARS, is achieved when one (or more) of the three frequencies ω_1 , ω_2 , or ω_3 is selected to be resonant with an electronic transition of the molecule under investigation (ω_1 and ω_2 are input laser frequencies, and ω_3 is the CARS frequency). When the electronic resonance condition is satisfied, enhancement factors as large as 400 have been observed. The OH concentration in flames is often as large as a few percent; however, the conventional CARS is difficult to detect because of the interference caused by the H₂O CARS spectrum which overlaps the OH CARS spectrum. Resonant CARS should be of considerable aid in this case.

The approach chosen in this program will be, first, to investigate thoroughly resonant CARS in the OH radical and, then, to assess the practical utility of resonant CARS as a viable diagnostic technique. Although laser-induced fluorescence is a useful diagnostic for OH in combustion research, it is an incoherent technique which may not be applicable to practical combustion devices with severely limited optical access, whereas the coherent CARS process is attractive because of the laser-like CARS beam which is generated. Moreover, the physics of how fluorescence quenching scales at pressures higher than one atmosphere is not yet established. The OH radical was chosen because of its extremely important role in combustion chemistry, and because so much is known about OH spectroscopy. In order to achieve electronic resonance and simultaneously satisfy the vibrational CARS condition ($\omega_1 - \omega_2 =$ Raman active vibration), both ω_1 and ω_2 must be generated from tunable, narrowband UV lasers. One of these UV sources is chosen as ω_1 (the pump), and will be tuned to be resonant with an OH electronic transition, and ω_2 (the Stokes frequency) will be scanned to generate the resonant CARS spectrum. Measurements are to be made in a uniform flame of high OH content.

During the first year of the contract, the two tunable narrowband dye lasers have been set up and tested. For each of these dye lasers, the tunability range has been measured, the laser linewidth estimated, and the output power optimized for frequency doubling in KDP crystals. One of the dye lasers is a commercial unit (Quanta-Ray PDL-1) and the second was constructed using the Littmann grazing grating configuration. Both dye lasers are driven by a frequency-doubled, Quanta-Ray pulsed Nd:YAG laser and have quite similar operating properties. The tuning range for each laser is more than 10 nm (for the fundamental) which is more than adequate to cover the OH CARS spectrum. The energy in each dye laser fundamental frequency is 15 millijoules. This energy is converted to 1.5 to 2.0 millijoules of UV by the KDP frequency doubling crystals. This should be more than enough energy for resonant CARS because previous experiments at UTRC demonstrated that saturation of fluorescence occurs at about 1 millijoule. The linewidth (before doubling) of each dye laser is about 0.3 cm^{-1} , quite suitable for selecting a single OH transition for resonance, and for performing CARS experiments. Within the past month, a Quanta-Ray WEX-1 (Wavelength Extender) has been installed at the output of the Quanta-Ray dye laser. The WEX functions to maintain constant power output from the frequency doubling crystal as the fundamental dye laser frequency is tuned. This is achieved by means of a feedback loop which angle tunes the crystal to maintain constant UV output power. It has been demonstrated that the WEX-1 can track the tuning of the PDL-1 with UV energies of 1 to 1.5 millijoules. The constant power output provided by the WEX is essential for scanning the Stokes frequency, ω_2 , in order to generate the CARS spectrum. Presently the dye lasers operate at wavelengths centered at 590 and 640 nm respectively. The laser dyes were so chosen to place ω_1 in the $0 \rightarrow 1$ (in absorption) band of the X $^2\Pi_{3/2}$ to A $^2\Sigma_{1/2}$ electronic transition of OH.

Two different burners, essentially the same in design, but differing in scale, have been tested for stability and uniformity. The burner design uses an alternating array of tubes and holes in a plate, the tubes and holes each fed separately from different plenum chambers, i.e. a closely-spaced multiple diffusion flame configuration. Both of these combinations produce very homogeneous flames (to the eye). The OH emission spectrum was easily observed and the 0-0 and 1-0 (in emission) bands easily observed. This design was chosen to avoid flashback when using fuel/oxidizer combinations such as H_2/O_2 or CH_4/O_2 . This type of burner was chosen over a premixed sintered porous plug burner because previous experience demonstrated that at atmospheric pressure erosion occurs with a H_2/O_2 flame, and clogging takes place for hydrocarbon flames.

The next step of this program will be to select the ω_1 frequency to be resonant with a single rotational line and then to search for resonant CARS by scanning ω_2 , holding ω_1 fixed on resonance. The test of resonance enhancement will be performed by tuning ω_1 in and out of resonance.

Interactions (Coupling Activities)

During this research period the following papers were presented:

1. "Resonant CARS Detection of OH Radicals" by J. F. Verdieck, R. J. Hall and A. C. Eckbreth was presented by J. F. Verdieck at the 1982 AFOSR Research Meeting on Diagnostics of Reacting Flow at Stanford University, February 25-26, 1982. This paper reviewed CARS activities at UTRC and gave the status of progress on the subject contract.
2. "Resonant CARS for Combustion Diagnostics" by J. F. Verdieck, R. J. Hall and A. C. Eckbreth was given at the 12th Northeast Regional Meeting of the ACS in Burlington, VT, June 27-30, 1982. This brief paper included an introduction to CARS and an outline of resonant CARS.
3. A seminar, reviewing CARS diagnostics, was given by A. C. Eckbreth at the AF Rocket Propulsion Laboratory on July 13, 1982. On July 14th, A. C. Eckbreth presented the "Technical Directors Seminar" on CARS diagnostics at the Naval Weapons Center, China Lake.

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