First Experimental Studies of the Resonant Optothermoacoustic Detection Technique

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Abstract: A novel technique for detecting optical absorption in gases is demonstrated. The technique called resonant optothermoacoustic detection (ROTADE) is based on energy transfer from the optical beam area to a mechanical oscillator via molecular diffusion.
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Optothermal detection is a spectroscopic technique where the energy input into gas or other media caused by absorption of optical radiation is measured directly by means of a thermal detector, usually using pyroelectric detectors or bolometers (see [1-3] and references therein). A fraction of the absorbed energy is transported to the thermal detector by heat conduction or molecular diffusion. In this work the conventional thermal sensor was replaced by a quartz tuning fork (QTF), and the optical power input into the gas was modulated at the QTF resonant frequency. We called this approach "resonant optothermoacoustic detection", or ROTADE. The physical principles of ROTADE sensor and its simple implementation are illustrated in Fig. 1.

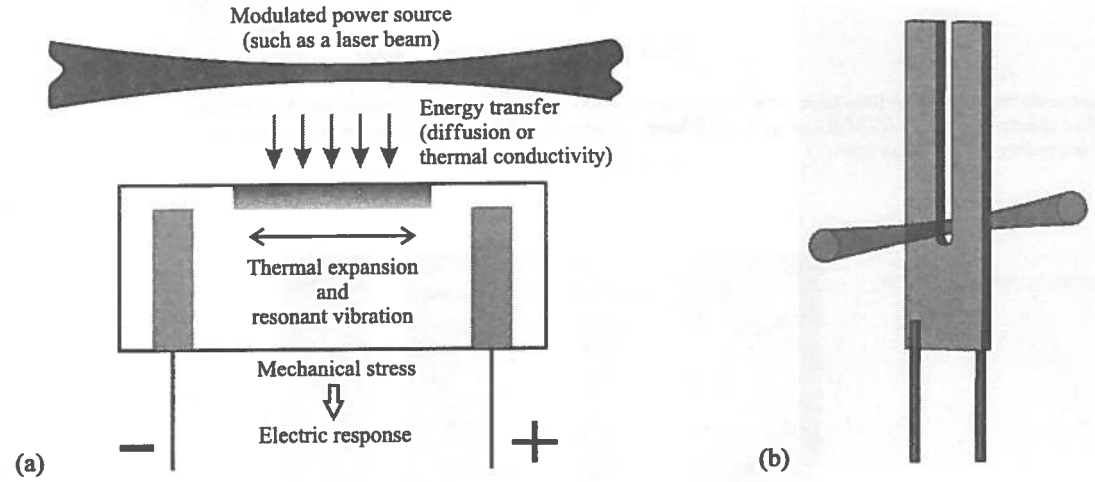

Fig. 1. (a) Principle of ROTADE sensor operation. Local thermal expansion of a piezoelectric crystal leads to the stress enhanced by mechanical resonance in the crystal. (b) One of the possibilities of practical implementation of the ROTADE technique using QTF.

Compared to a conventional optothermal spectroscopy, ROTADE allows the sensor to operate at a higher modulation frequency, thus reducing 1/f noise. The same experimental setup can be used to conduct quartz enhanced photoacoustic spectroscopy (QEPAS) [4], which is closely related to ROTADE. QEPAS relies on energy transfer from the initially excited molecular vibrational state to the translational degrees of freedom. In some cases this process is too slow to follow the 32.8 kHz modulation required for QEPAS. In other cases, the resonant energy transfer can result in vibrational excitation of nitrogen, which relaxes very slowly. ROTADE, on the other hand, detects the energy delivered by molecules even if this energy is still in the form of vibrational excitation. The molecules will then release their energy to the QTF upon collision with its surface.

In a simple sensor configuration shown in Fig. 3(b), both QEPAS and ROTADE signals are present, but one of them is dominating depending on the laser waist position, gas pressure, and V-T relaxation time. While the acoustic wave driving QEPAS decays slowly and has a wavelength of ~1 cm, the diffusion wave driving ROTADE decays much faster. Its wavenumber κ is determined by two equations given in Ref [5]

technology rather than expensive and less sensitive MIR cameras or microbolometers. We expect that a combination of VIPA optical alignment improvements and upconverted comb tooth filtering will allow the comb modes to be individually resolved.

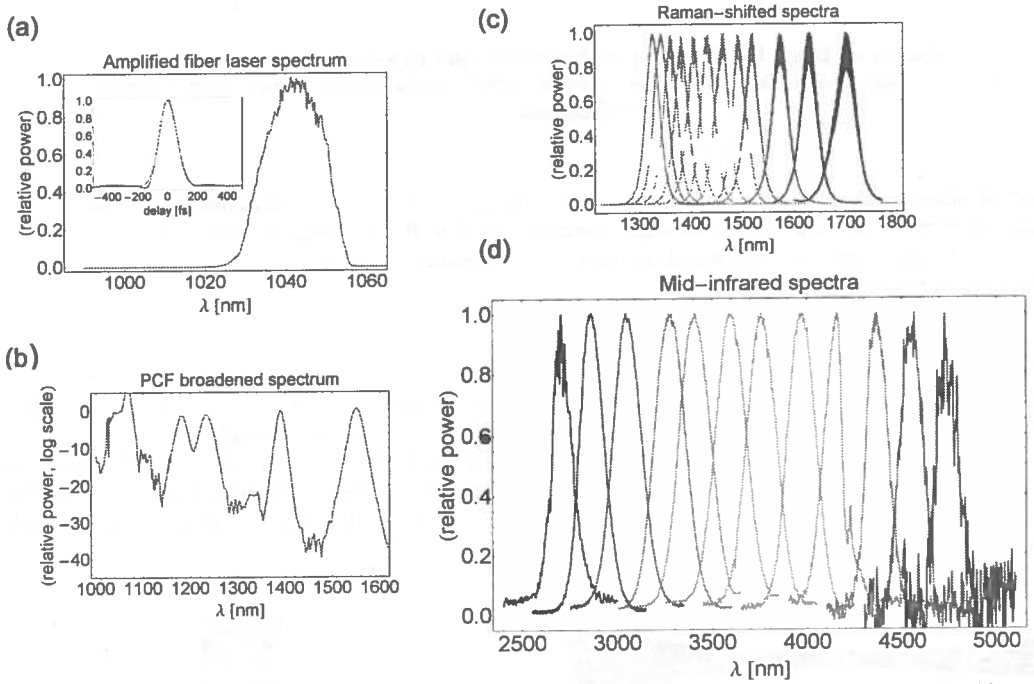

Fig.2. (a) Spectrum from amplified fiber laser, with inset showing ~130fs pulse duration. (b) Broadened spectrum. (c) Raman shifted spectra from PCF. (d) MIR spectra from difference frequency mixing between the amplified laser spectrum and the corresponding Raman peaks from (c).

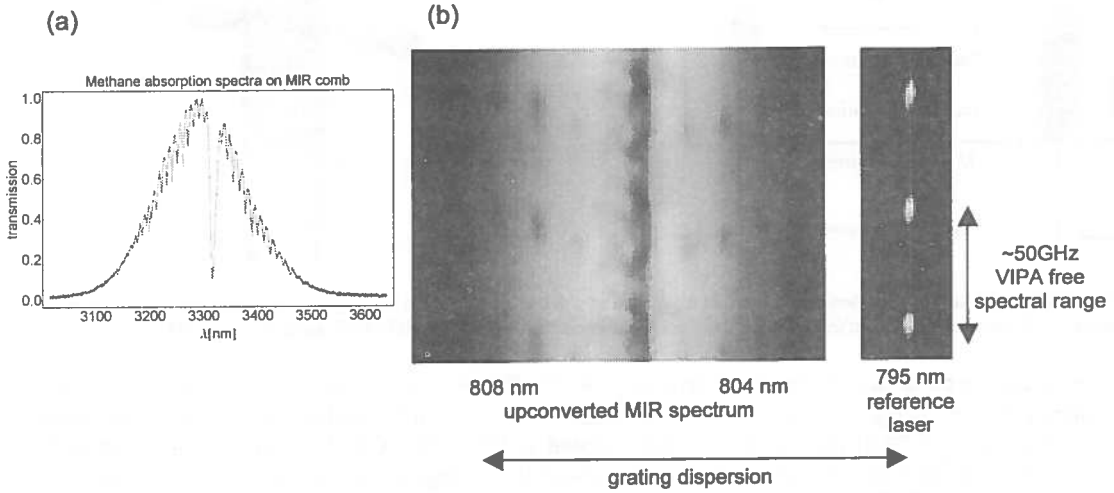

Fig. 3. (a) Monochromator trace of methane transmission with MIR comb. (b) CCD image of the upconverted spectrum using a VIPA and grating spectrometer. The dark central band corresponds to the strong absorption of methane near 3310nm, and the dark spots match the evenly-spaced weaker absorption features of part (a). Present resolution is ~ 8GHz.

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$$\kappa = \frac{1+i}{L(\omega)}, \quad L(\omega) = \sqrt{\frac{2D}{\omega}\frac{760\ \text{Torr}}{P}} \tag{1}$$

where $\omega=2\pi f_0$, f_0 = 32.8 kHz for the QTF, P is the gas pressure, and D is a diffusion coefficient at 1 atm. We performed measurements with CO_2 gas, where D=0.12 cm^2/s [6], and at 20 torr we calculate L=67 μm, and hence the wavelength $\lambda_{th}\approx$420 μm.

Experimental investigations of ROTADE and its comparison with QEPAS were performed using a near-infrared diode laser at 1.57 μm to probe the CO_2 absorption line at 6361.25 cm^{-1}. A fiber collimator and a refocusing lens were used to focus the laser to a ~15 μm diameter waist. Its position was scanned in the QTF plane using a 3D translation stage with computer-controlled actuators. The signal was acquired in 2f wavelength modulation mode, similar to earlier QEPAS publications. The transmitted laser power was measured in order to obtain an outline of the QTF. Fig. 2 shows the experimental results.

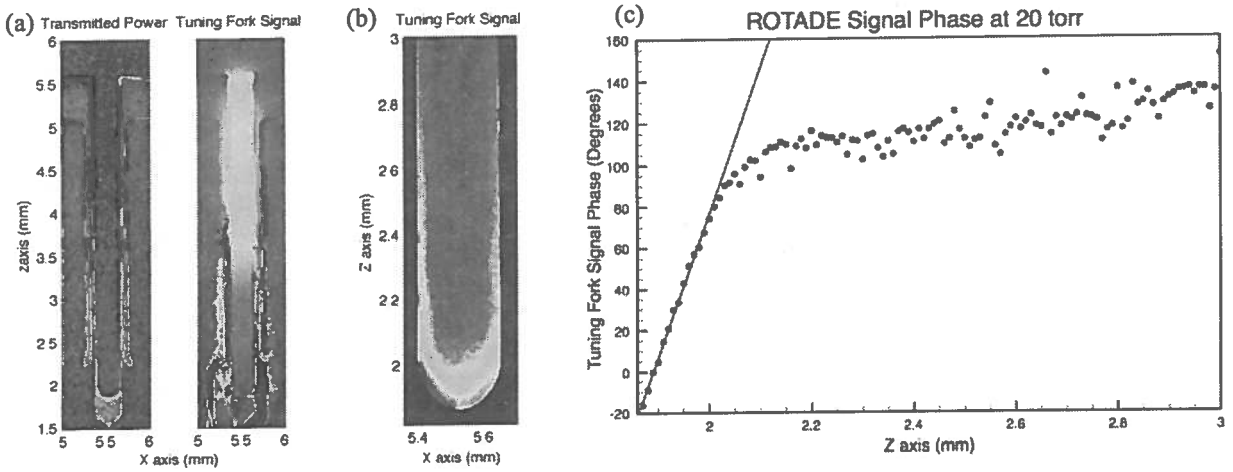

Fig. 2. (a) The image of the tuning fork depicts the transmitted power and signal as a function of beam position for 300 torr of pure CO_2. The observed signal is due primarily to the photoacoustic effect. (b) The image of the tuning fork shows the signal as a function of beam position at a reduced pressure of 20 torr of pure CO_2. The tuning fork has been colored black to highlight the area where the ROTADE effect can be observed. (c) The plot shows the phase of the tuning fork signal as a function of the laser focus position on the z-axis centered at 5.5 mm on the x-axis (QTF plane of symmetry) at 20 torr of pure CO_2.

Based on the slope of the initial part of Fig. 2c plot and using a flat thermal wave approximation, the self-diffusion coefficient of CO_2 at atmospheric pressure was calculated to be 0.12 cm^2/s, in agreement with published data [6].

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