ABSTRACT
In the present study the possibility to apply time resolved Laser Induced Thermal Grating Spectroscopy (LITGS) to detect fuel concentration and temperature in mixtures and flames at atmospheric pressure or higher is investigated. The resonant IR single photon absorption of two short pulse pump beams is used to initially generate a population grating, decaying into a thermal grating due to relaxation processes in the gas mixture. The thermal grating evolution is followed by monitoring the scattered signal of a cw visible probe beam after the end of the pump pulse. The use of the IR optical transition of diesel fuel assured a high species selectivity and a negligible influence of the visible emission background due to the presence of electronically excited species in flames. Fuel concentration and temperature measurements in a pressurized cell, with pressure ranging between 0.1 and 1.5 MPa, and in a diffusion turbulent flame generated by a burner feed with diesel fuel operating at atmospheric pressure are presented. The experimental investigation shows that LITGS signal increase linearly with gas density. This characteristic makes LITGS a very interesting technique for fuel distribution and temperature measurements in hostile (high-pressure and turbulent flow) environments. Detection limit for diesel fuel at atmospheric pressure is found to be about 40 ppm and it decreases with the increase of the pressure. The extremely low reachable detection limit makes this technique suitable also for monitoring minor species and radicals.

INTRODUCTION
The knowledge of temperature and chemical species concentration in the combustion processes is of notable importance in combustion diagnostic. In fact, many minor species and radicals could be observed only directly during combustion, being their lifetime extremely short. Many techniques have been applied to combustion diagnostic but almost all of them present significant limits when applied to cases of practical interest (e.g. gas turbines and I.C. engine combustion chambers, etc.) because of the simultaneous presence of high pressures and temperatures. In fact, Coherent Anti-Stokes Raman Scattering (CARS), Laser Induced Fluorescence (LIF) and Degenerate Four Wave Mixing (DFWM), which are among the most widely used techniques for combustion diagnostic when employed to high pressure applications show strong limits due to the higher importance of the non-resonant background in CARS, to the collisional quenching effect in LIF, and to pressure broadening in DFWM [1, 2]. Currently new optical diagnostic techniques based on time resolved evolution study of thermal gratings are under development. These techniques are particularly indicated for temperature and species concentration (NO, NO2, OH, etc.) [3, 4, 5, 6] measurements in hostile environment because they seem not to suffer signal deterioration due to high pressures. This characteristic makes them particularly suitable for engine and high-pressure burner applications. Their signal is generated by the modulation of a read out laser beam, from hereafter referred to as "probe beam", scattered by a grating formed by the interference of two different laser beams referred to as "pump beams". The interference causes a periodic variation of the energy intensity distribution that corresponds to a variation of the index of refraction of the medium in which the grating is formed. The variation in the space of the electric field in the region of interference produces a transfer of mass toward the regions to higher intensity generating a sound wave that is propagated along the two perpendicular directions to that of the interference fringes. In this way a grating whose characteristics vary in time according to the physical parameters of the surrounding medium is generated. If the frequency of the pump beams is in resonance with some of the chemical species presents in the interference region, a thermal grating generated by a
thermalization process of excited molecules via inelastic collisions, predominantly through V-T relaxation, with a redistribution toward the kinetic energy of all the particles. If resonance conditions are not verified the thermal grating could only be generated by electrocostriction or black body radiation absorption. In general, the gratings generated by thermalization processes persist longer and present a higher contrast. Both characteristics contribute to improve signal quality and intensity, thus reducing the signal/noise ratio and, therefore, the measurements uncertainty.

The purpose of the present investigation is to prove the applicability of the LITGS technique to temperature and fuel concentration measurements in turbulent diffusion flames.

BACKGROUND THEORY

When two laser beams of wave length $\lambda$ and equal polarization cross each other with an angle $2\theta$, they interfere giving a grating whose intensity varies in the space. If the interaction of two electromagnetic plain and monochromatic waves of the form of Eq (1) is examined the resulting electromagnetic intensity field is given by Eq. (2).

$$E_j = A_j \exp(i\vec{k}_j \cdot \vec{r})$$  \hspace{1cm} j = 1, 2  \hspace{1cm} (1)

Where $A_j$ is the wave amplitude, $\vec{k}_j$ is the wave vector defined as $2\pi/\lambda$ and $\vec{r}$ is the spatial coordinate,

$$I(\vec{r}) = \frac{1}{2} [E_1(\vec{r}) + E_2(\vec{r})][E_1(\vec{r}) + E_2(\vec{r})]^* =$$

$$= \frac{1}{2} \left[ |A_1|^2 + |A_2|^2 + 2|A_1||A_2|\left(1 - 2\sin^2 \theta\right)^{\frac{1}{2}} \cos(\vec{k}_2 \cdot \vec{k}_1) \cdot \vec{r} \right]$$  \hspace{1cm} (2)

Where $p=0$ and $p=1$ for parallel and orthogonal polarization with respect to the plane defined by the wave vectors $\vec{k}_1$ and $\vec{k}_2$, respectively. The interference grating spatial wavelength $\Lambda$ is related to the angle $2\theta$ between the two pump beams, by Eq (3) directly derived by Eq (2).

$$\Lambda = \frac{2\pi}{|\vec{k}_1 - \vec{k}_2|} = \frac{\lambda}{2\sin \theta}$$  \hspace{1cm} (3)

Eq (3) shows that $\Lambda$ reaches its minimum value of $\lambda/2$, for $\theta=90^\circ$. An analysis of Eq. (2) shows some interesting grating features. In the first case, the maximum grating intensity value in independent on the crossing angle between the two pump beams. Consequently, the energy transferred to molecule does not depend on $\theta$. If the quality of the grating is estimated by the contrast index $V$, defined as

$$V = \left( \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}} \right)$$

where $I_{\text{max}}$ is the intensity on the grating crest and $I_{\text{min}}$ is that on the grating valley, it could be observed that in the ideal case, in which $I_{\text{min}} = 0$, $V$ reaches its maximum value of $V=1$. From Eq. (2) and from the definition of the contrast index is seen that the grating quality also depends on the direction of the incident wave polarization. In fact, being $\sigma$ and $\pi$ the geometries in which the polarization vector results to be respectively parallel and orthogonal to the plane of the wave vectors $\vec{k}_1$ and $\vec{k}_2$, two different cases are possible: the first in which beam intensities are the same and the second in which they are different.

Figure 1a: Contrast index plot against beams crossing angle for orthogonal polarization.

Figure 1b: Contrast index plot against beams intensity ratio $A_1/A_2$.

In the first case for polarization of type $\sigma$, corresponding in Eq. (2) to a value of $p=0$, the contrast index, $V$, engages his maximum value, $V=1$, for each value of $\theta$ and, therefore, for each $\Lambda$. Whereas, for polarization of type $\pi$, corresponding in the Eq. (2) to a value of $p=1$, the law of variation of the
contrast index is shown in Figure 1a. Figure 1a shows that in this case good grating contrast are obtained only for $\theta=0^\circ$ e $\theta=90^\circ$, whereas for $\theta=45^\circ$ waves $E_1$ and $E_2$ are perpendicular, thus none interference path is generated. Similar conclusion could be reached for the second case. However, it is interesting to note, Figure 1b, that beams intensity ratio has only a mild influence on grating quality. Thus, beams intensity differences up to 20% could be observed with no meaningful reductions of the contrast index, $V$.

The presence of an interference grating originates energy exchanges between the gases present in the intersection zone of the pumping beams, that produces a variation of the optical susceptibility of the gaseous medium in which the measure is carried out. Such a variation regards, in particular, the not linear third order component $\chi^3$. Being the susceptibility a complex quantity $\chi=\chi^R+\chi^I$, different types of gratings are prevalingly generated if either its real or imaginary parts are mainly involved in the process.

The real component gives rise to the so-called phase or refractive index gratings including sound waves generated by opto-acoustic effect, electrocostriction and thermalization, due to redistribution of vibrational energy in thermal energy (LITGS), effects. In these cases is possible to study the temporal evolution of the formed grating by analyzing the modulation with which a third beam of arbitrary wavelength, $\lambda_{ac}$, is coherently scattered by the grating. However, scattering only occurs when the reading beam sees the grating under such an angle that the Bragg reflection condition is satisfied.

The imaginary component is responsible for the so-called amplitude gratings, due to not linear processes with coherent light scattering where the signal generation is determined by the wavelength matching. Amplitude gratings include population and polarization gratings. From a solution of the linearized Navier-Stokes equation under the hypothesis of $\text{Re} = a\Lambda/\nu >> 4\pi^2$, where $a$ is the speed of the sound and $\nu$ the kinematics viscosity, Daney et al. [7] and Dreizler et al. [8] have shown that thermalization effects are preponderant in high density environments, whereas in low density environments are dominant the effects due to overpopulation of the molecules upper energy levels. For measurements at atmospheric pressure and temperatures of the order of 1500-2000 K both effects are comparable.

Another very important parameter to evaluate grating quality is its scattering efficiency. $\eta$, defined as the ratio between the intensity of the signal, $I_{\text{sig}}$, and that of the probe beam, $I_{\text{pr}}$.

$$\eta = \frac{I_{\text{sig}}}{I_{\text{pr}}} = \left(\frac{L\Delta \alpha}{4}\right)^2 + \left(\frac{\pi L \Delta n}{\lambda_{\text{pr}}}\right)^2 \quad (4)$$

Where $\Delta \alpha$ and $\Delta n$ are the difference of the value of the absorption coefficient and of the index of refraction between the crests and valleys of the grating, respectively, and $L$ is the grating length, that results to be about the length on which the two pump beams interact.

The first term of Eq. (4) represents the contribution due to population grating, whereas the second term expresses the thermalization effect. From the Eq. (4), it is seen that the signal does not directly depend on the intersection angle, $2\theta$, between the two pump beams, but only indirectly, being the interaction zone proportional to $L = 2w_0/\text{sen} \theta$ where $2w_0$ is the pump beams diameter. The thermalization process causes fluctuations of the index of refraction, $n = \sqrt{\varepsilon}$, of the gas present in the measuring volume, where $\varepsilon$ is a tensor defined as $\varepsilon = 1 - \chi$. Such fluctuations are described by the following equation:

$$\Delta \varepsilon = \left(\frac{\partial \varepsilon}{\partial \rho}\right)_T \Delta \rho + \left(\frac{\partial \varepsilon}{\partial T}\right)_\rho \Delta T \quad (5)$$

Where density perturbation, $\Delta \rho$, is the following function of the total pressure and of the entropy of the system:

$$\Delta \rho = \left(\frac{\partial \rho}{\partial \varepsilon}\right)_S \Delta \varepsilon + \left(\frac{\partial \rho}{\partial S}\right)_\rho \Delta S \quad (6)$$

The first term of the right hand side of the Eq. (6) represents the contribution of density adiabatic perturbations and, therefore, it describes the effect of opto-acoustic waves, whereas the second term describes the isobaric variations and, therefore, the thermalization grating. The grating relaxation time depends on mass, energy and momentum transport phenomena within the measuring volume. Thus, opto-acoustic contribution to the grating is dumped, primarily because of the gas viscosity $\mu$, with the following characteristic time constant:

$$\tau_{ac} = \left(\gamma - 1 + \frac{4\mu c_p}{3\lambda_{\text{term}}}ight) \frac{\Lambda^2 \rho c_p}{2\pi^2 \lambda_{\text{term}}} \quad (7)$$

Where $\gamma$ is the ratio between the specific heat at constant pressure, $c_p$, and volume, $c_v$. $\lambda_{\text{term}}$ is the gas thermal conductivity. On the other hand, thermal grating contribution, given by the second term of Eq. (6), decay with a characteristic time constant given by:

$$\tau_{\text{term}} = \frac{\Lambda^2}{4\pi^2 D} \quad (8)$$
Where $D = \frac{\lambda_{\text{term}}}{\rho c_p}$ is the gas thermal diffusivity. Therefore, grating time evolution is described by:

$$I(t) = C_1 \exp\left(-\frac{t}{2\tau_{ac}}\right) \sin(\omega t + \phi) + C_2 \exp\left(-\frac{t}{2\tau_{\text{term}}}\right)$$

(9)

Where $C_1$, $C_2$ and $\phi$ are constants to be determined experimentally and

$$\omega_s = \sqrt{\gamma RT / \Lambda}$$

(10)

is the frequency of the opto-acoustic oscillations.

**EXPERIMENTAL RESULTS AND DISCUSSION**

Preliminary measurements to find out LITGS detection limits for diesel fuel and to calibrate the technique were carried out in a pressurized vessel by using the experimental setup shown in Figure 2.

The pumping source was a CO$_2$ laser operating at 9.586 $\mu$m with a typical output energy of 1.3 J/pulse at 1.0 Hz, with a FWHM of 100 ns and a beam diameter of about 15 mm. The output beam, focused by a mirror with a focal length of 20 m, was passed through a beam splitter and divided into two pumping beams having energy of about 635±10 mJ/pulse. One of the pumping beams was routed through a delay line to equal the optical path of both the pumping beams prior to cross each other in the sample volume. The crossing angle was of 4°. After the sample volume one of the CO$_2$ beams was dumped whereas the other was redirected to a fast photon drag I.R. detector located inside a Faraday cage, to generate the trigger signal for the data acquisition system.

The use of a Faraday cage was necessary to cut off the electromagnetic noise generated by the CO$_2$ laser discharge process. The evolution of the grating was read by a probe beam generated by a cw He-Ne laser operating at 632.8 nm with a diameter in the scattering region of 1 mm and a measured power output of 16 mW. The probe beam was incident the sample volume under the Bragg angle $\Phi$ given by $\sin(\Phi) = \frac{\lambda_{pr}}{2\Lambda}$.

To reduce scattering from the undeflected portion of the He-Ne beam, it was necessary to pass the deflected beam through a diaphragm and a spatial filter before entering the photomultiplier. All signals were acquired by a digital storage oscilloscope and subsequently transferred to a personal computer for data analysis.

In a 200 mm long cell closed at its ends with 50 mm diameter ZnSe, coated with antireflection layers, windows preliminary measurements were made in order to calibrate the technique. Then temperature of turbulent diesel fuel flames where measured at different points in the flame and under different operating conditions of a 9.2 kW burner.

The use of I.R. pumping beams is motivated by the presence in the diesel fuel of many optical transition in the region between 9 and 13 $\mu$m as is shown in Figure 3.

![Figure 2: Schematic of the experimental setup](image1)

![Figure 3: Diesel fuel emission spectrum. Fuel density $\rho$=828 kg/m$^3$](image2)

Detection limit at atmospheric pressure and ambient temperature for diesel fuel (C$_{16}$H$_{29}$) is of only 40 ppm, and at a pressure of 1.5 MPa it becomes less than 20 ppm.

Figure 4 shows a typical recorded signal trace acquired during the calibration procedure. In the upper panel of Figure 4 signal oscillations around its mean value are plotted. It can be seen that signal intensity decreases exponentially with a time constant $\tau_{ac}$=3.47 $\mu$s. From the high frequency oscillations period it is possible to measure the speed of the sound by using Eq. (10), being the time $\tau_s$ between two subsequent signal peaks related to the oscillations frequency by the following relationship $\tau_s=1/\omega_s$. It is interesting to notice that at low total
pressure values, part of the signal is generated by electrocostriction gratings. This effect is visible at the beginning of the signal trace of the second panel of Figure 4 in correspondence of a time of 5 μs.

Figure 4: Typical LITGS signal. Proceeding from top to bottom are plotted: a) signal oscillations around the mean value, b) signal trace, and c) CO2 laser pulse, respectively. Operating condition: 0.2 Mpa and 293.5 K

Electrocostriction signal has been shown [6, 9, 10], to generate an oscillating signal whose period $\tau$ is equal to 0.5 $\tau$, and therefore is easily to account for its contribution. The influence of the electrocostriction signal on the overall signal is found to strongly decrease with gas density and it was not detectable at a pressure of 0.6 Mpa and ambient temperature. As pointed out by Paul et al. [11] the second term in Eq. (5) is quite small compared to the first, therefore only the dependence of the LITGS signal on the pressure was investigated.

To quantify the effect of total pressure changes and of fuel partial pressure on the LITGS signal a two step calibration procedure is followed in the present investigation. In the first step a fuel vapor concentration of 300 ppm is held constant while the total pressure is varied by adding nitrogen as buffer gas between 0.1 Mpa and 1.5 Mpa at a constant temperature of 293 K. The fuel vapor concentration of 300 ppm was chosen because it was the maximum fuel concentration achievable at a temperature of 293 K and a total pressure of 1.5 Mpa, being the fuel vapor tension at 293 K equal to 0.5 kPa. Figure 5a shows the obtained results. Symbols represent experimental measurements while the solid line is the linear regression to the experimental data. Figure 5a shows a linear dependence of the signal on total pressure, such a dependence was also observed by other researchers [6, 12], and it could be explained with the reduction of the molecules mean free path with pressure increase. This diminution provokes a faster thermalization of the exited molecules V-T energy thus increasing grating reflectivity by increasing the $\Delta n$ term in Eq. (4). In a second step, keeping a constant temperature of 293 K and a total pressure of 0.1 Mpa, measurements are carried out by varying the fuel vapor concentration changing its partial pressure between 0 and 0.5 kPa. The obtained results are plotted in Figure 5b. A linear dependence of the signal on vapor fuel partial pressure is observed. This dependence is justified by the greater energy per unity of mass absorbed in the grating region because of the higher absorbing molecules number density. Therefore a saturation value of the signal intensity is expected to be found at higher vapor fuel concentration but it has not been observed in the present investigation.

Figure 5a: Dependence of the LITGS signal on total pressure for a vapor fuel concentration of 300 ppm.

Figure 5b: Dependence of LITGS signal on partial vapor fuel pressure for a total pressure of 0.1 MPa

To combine the information acquired during the calibration procedure in a single calibration surface the three following considerations are made. First of all, when the partial fuel pressure or the total pressure is zero also signal intensity has to
be equal to zero; second, the intersections of the calibration surface with planes at constant total pressure or constant fuel partial pressure have to be linear; third, the reasons why a variation of the LITGS signal with pressure for the cases of Figure 5a and 5b are physically independent and therefore their contribution act simultaneously. The obtained calibration surface with iso-signal contours is reported in Figure 6.

**Figure 6: LITGS signal dependence on pressure.**

**BURNER RESULTS**

Calibrated the system, experimental measurements on a small power (9.2 kW) burner able to produce turbulent diffusive flames at atmospheric pressure were carried out. Work at atmospheric pressure it means to test the technique under not optimized conditions due to the low intensity of the obtainable signal, as shown in Figure 6, that in some of the investigated condition becomes even comparable with the noise. The fuel is injected under steady state conditions by a nozzle located in the bottom part of the burner and the spray axis is directed vertically in the same direction of the burner axis. Under of the nozzle a plenum with three ports directing the primary combustion air flow 30° from the burner radius and 30° upward to provide swirl is located. The primary combustion air is provided by the laboratory compressed air system via a 40 liter slightly pressurized vessel to ensure a constant feeding pressure and a rotameter to measure the supplied primary combustion air flow. Measurements at different locations (between 5 mm and 28 mm from the burner exit) along the flame axis were carried out and their results are here presented. Measurements between 30 and 42 mm from the burner exit were also made, but because of the higher temperature present in this region and of the low fuel concentration (most of the fuel is already burned), the signal was very noisy and accurate measurement were not possible. In general the following trends are observed.

First, signal noise increases moving from the burner exit due to the presence of higher density gradients. Second, the intensity of the LITGS signal at the beginning grows with the axial distance from the burner exit following the gas temperature growth that increase the fuel vaporization rate, then it suddenly drops owed to the decrease of fuel vapor concentration due to the combustion process. Third, in the high temperature zones, the low-density values allow the contemporary presence of thermal and electrocostriction gratings. In Figures 7a and 8a, typical LITGS signals at 28 mm an 5 mm, respectively from the burner exit are reported. Operating conditions are: fuel flow rate 0.23 g/s and air flow rate 2.0 g/s.

**Figure 7a: LITGS signal acquired at 28mm from the burner exit, as roughly shown by the inset figure, in the region of fluctuating flames.**

**Figure 7b: FFT of the LITGS signal plotted in Figure 7a.** With arrows are indicated the peaks due to the LITG and the electrocostriction signals. The high frequency noise region is also shown.
The first thing to notice is that in Figure 7a the signal presents a much higher noise. A possible explanation is the presence of density gradients due to relatively high dimension turbulence eddies and to combusting droplets. These not uniformly distributed density gradients, in fact, divert both the two pump and the probe beams varying grating characteristics and reflection efficiency.

In Figure 7a are indicated by arrows the first, the third and the fifth peak of the signal generated by electrocostriction grating. The correspondent beats of even order (second, fourth and sixth) are not visible because they coincide with the peaks of the LITGS signal, being $\tau = 0.5\tau_s$. The frequency analysis of the signal of Figure 7a is reported in Figure 7b where are clearly visible the oscillation frequency of the LITGS signal, at 0.62 MHz, and that of the electrocostriction at a frequency double with respect to the first, at 1.24 MHz. In Figure 8a a typical LITGS signal at 5 mm from the burner exit is plotted. In this case, the signal is more clearly identifiable and electrocostriction signal is much less. From a statistic analysis of acquired data (at least 20 single shot acquisitions) a temperature of $1380\pm250$ K and $340\pm40$ K for the positions of Figure 7a and 8a, respectively are obtained. The relatively high error magnitude is mainly due to the reason discussed before. However flame fluctuations also contribute to magnify the error of the measurements made at 28 mm from the burner exit. The error due to the uncertainty of the gas composition, required to calculate the mixture gas constant $R$ in Eq. (10) in the measuring point, could be neglected. In fact, differences in the value of $R$ for real conditions with respect to the value of $R$ calculated assuming chemical equilibrium for overall lean combustion is much less than 1%. The measured temperatures ($1380\pm250$ K and $340\pm40$ K) are in good agreement with those measured by means of spectral analysis of flame radiation.

Using the calibration diagram of Figure 6, the fuel concentration at the different locations was also evaluated. A statistical analysis of the acquired data (at least 20 single shot measurements) shows vapor fuel concentration values of $8650\pm10\%$ and $4870\pm5\%$ ppm for the conditions of Figure 7a and 8a, respectively. The error, even though is less than that obtained for temperature measurements, it is still dependent mainly on density gradients due to turbulence and to burning droplets. The reason why turbulence is less effective in determining the magnitude of the measurement error is that the grating efficiency does not change with the pumping beam crossing angle as Eq. (4) shows. Thus, in the short time needed to record the amplitude of the first signal oscillation turbulence perturbations, are minimal. For concentration measurements, therefore, turbulent fluctuations are found mainly to affect the shot to shot instantaneous fuel concentration in the measuring point and to have some effect in steering the probe beam. Experimental data acquired with different techniques are not available. Therefore, in the spirit of speculation, to assess LITGS reliability in measuring fuel concentration, measurements in a not combusting laminar air flow seeded with 5000 ppm of C$_2$H$_6$ were made. To be in resonant condition with the ethane molecule the CO$_2$ laser was operated at a frequency of 9.603 $\mu$m and the experimental setup was accordingly modified. The concentration measured with LITGS technique was of 4978$\pm$185 ppm thus confirming its extremely good capability.

**CONCLUSIONS**

The investigated technique has been proven to be a very promising tool for combustion diagnostic in particularly hostile environments. For the first time LITGS has been successfully applied to temperature and diesel fuel concentration measurements in atmospheric turbulent diffusive
flames. The signal intensity was found to be linearly dependent on both total pressure and partial pressure of the investigated species. Because of such dependence measurements made in the flame high temperature regions were characterized by the greater errors. Furthermore, measurements in the high temperature regions, because of the accordingly low density, have shown the rise of electrostriction grating whose signal had to be taken into account in the interpretation of the experimental data. In general, temperature measurements were very precise and of immediate determination, depending only on the oscillation period $\tau$ of the opto-acoustic waves, while concentration measurements required an adequate calibration of the technique to be able to correctly interpret experimental data from in flame measurements. The effect of turbulence, burning droplets and soot particles on LITGS signal resulted in a high frequency noise source that was possible to filter by a FFT analysis of the acquired data. From the present investigation it is possible to conclude that LITGS for its experimental simplicity is, for low pressure measurements, an attractive alternative to more classic combustion diagnostic techniques as LIF and CARS, while it opens new interesting perspective to high pressure combustion diagnostic.

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