Saturation curves of two-color laser-induced incandescence measurements for the investigation of soot optical properties

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Abstract

Two-color laser-induced incandescence measurements are carried out in diffusion flames and at the exhaust of a home-made soot generator, both fuelled with ethylene and methane. Two-color prompt LII signals, their ratio and the corresponding temperature have been investigated as a function of laser fluence. LII spectral measurements have also been performed in all conditions for validation. In order to gain information on the soot absorption properties the effects of fuel, soot load and gas/particle initial temperature on LII measurements have been investigated. The results suggest that the incandescence signal is sensitive to both optical and non-optical physical properties, and therefore the understanding of the LII response to these properties is mandatory for a correct interpretation of the LII measurements. Such issues can be overcome working at high laser fluences, where the saturation curves show independence on the experimental conditions if the soot absorption function near soot sublimation threshold is known.

1. Introduction

Fine and ultrafine particulate matter (PM), essentially generated by combustion systems, are currently of great interest due to their effect on human health and climate change. The effects are strictly connected with the chemical composition as well as the size, density and shape of the particles. The need to measure and characterize carbonaceous particles, both inside and at the exhaust of combustion systems as well as in the environment, has triggered the interest of the scientific community to develop and apply advanced diagnostic techniques in different experimental conditions. The laser-induced incandescence (LII) technique is a widely used diagnostic tool to measure carbonaceous particles concentration and dimension with in-situ, time and spatially-resolved measurements [1-3]. In particular, the two-color LII technique is attractive as it returns absolute soot concentration measurements with an optical calibrated arrangement, without the need of calibrated source, which could be a problem in the case of very low concentrations [4-13]. Nevertheless, the interpretation of the incandescence signal as well as of other soot optical diagnostic measurements requires the knowledge of the optical properties of the carbonaceous particles under investigation. Many studies on the optical properties are reported in the literature. Several investigations about the soot absorption function, E(m), have been performed both theoretically and experimentally, resulting in a wide spread of values and consequently in large discrepancies and uncertainties in the measurements of temperature and particles concentration. These values have been obtained in different experimental conditions, in diffusion and premixed flames as well as in the overfire region of flames. Different behavior of E(m) values with wavelength has been found in the literature, resulting in increasing, decreasing or even constant values with wavelength [14-20]. The wide spread of the refractive index values and consequently of the absorption function E(m) is strictly related to different fuels, the diagnostic tool implemented and soot composition, such as the hydrogen to carbon ratio [13], which in turn depends on the different nature and aging stage of the carbonaceous particles. In fact, the presence of nascent and mature soot particles in flames, characterized by different nano-scale carbon organization [21-25], which could be responsible for different optical properties, has already been shown in the literature. Moreover, soot maturity and composition could affect not only the optical properties but also some other physical properties which, in turn, could affect the laser-induced incandescence signals [26 and reference therein]. Examples of these physical parameters could be the aggregate reconstruction and collapse after particle coating, a decrease of the surface roughness due to increasing soot maturity and a change in the graphite surface when hydrogen is bonded to the surface. Therefore, in order to correctly interpret the LII measurements, a better understanding of the LII response to these properties is mandatory.

In this work two-color laser-induced incandescence measurements are performed in a widely studied ethylene/air and methane/air co-flow diffusion flames of 6.5 cm height as well as at the exhaust of a home-made soot generator. This last device consists essentially of a quenched diffusion flame, where the choice of the fuel, its flow rate and the height of the Nitrogen quencher allow changing the overall soot load [27]. In this work the soot generator is fed with the same fuels as the laboratory flames under analysis, which means ethylene and methane. In flames measurements are carried out along the flame axis and on the external axial symmetric soot layer in order to overcome self-absorption effects. Correspondingly, the analysis at the exhaust of the soot generator has been performed by quenching the soot generator flame at the same heights as for the in-flames measurements. The aim is to explore the use of LII as a tool to gain information on the nature and optical properties of soot particles in different experimental conditions and in a wide range of soot load. Moreover, the dependence of the absorption properties on particle temperature and consequently the effect of the initial particle temperature on the incandescence temperature value have also been considered. For this reason the investigation on in-flame and cold nanoparticles, as produced at the exhaust of the soot generator, is carried out.

Two-color and spectral laser-induced incandescence measurements have been performed by varying the laser fluence. In this way, from the ratio of the incandescence signals detected over the two spectral regions the incandescence temperature as a function of laser fluence can be derived, which in turn provides information about the absorption properties of the carbonaceous particles under investigation.

Although at high laser fluences the plateau regime is reached independently from the source of the soot investigated, at low laser fluences some differences in the overall behavior of the incandescence temperature are observed. This could be explained by a combination of different phenomena, such as the initial temperature of in-flame and cold soot particles [28], the presence of coating on the soot particle surface [29], a likely difference in the inner structure, density and heat capacity of the particles under investigation as well as in the absorption properties.

2. Experimental set-up

The laser-induced incandescence experimental apparatus is shown in Figure 1. The fundamental beam of a Nd:YAG laser (1064 nm, Quantel, Big Sky CFR 400, Δt= 7 ns, top-hat output) running at 5 Hz was used as excitation source. The laser beam was steered onto an aperture, and then imaged onto the probe volume with a magnification of 1. Such configuration allows us to produce a top-hat, which means quite uniform and sharply edged, laser beam cross-section in the probe volume [4, 30]. Due to the wide range of soot concentration investigated (more than one order of magnitude), the aperture diameter was properly changed accordingly to the condition under investigation, though maintaining the laser cross section and the soot concentration in the probe volume as uniform as possible. In particular, apertures of 1 mm and 2 mm for ethylene, and of 1.5 mm and 3.5 mm for methane were used for the flame and the soot generator, respectively. For ethylene flame the aperture has been chosen to have a reasonable spatial resolution in order to resolve the annular soot layer. For methane flame a higher aperture was necessary due to the lower soot load with respect to the ethylene flame. For cold soot measurements, the considerably lower soot load due to Nitrogen dilution requires the use of a larger beam aperture with respect to the in-flame soot. The top-hat distribution of the laser intensity has been verified for all apertures; an example is inserted in Fig 1. For each experimental condition neutral density filters were used to obtain the LII saturation curve.

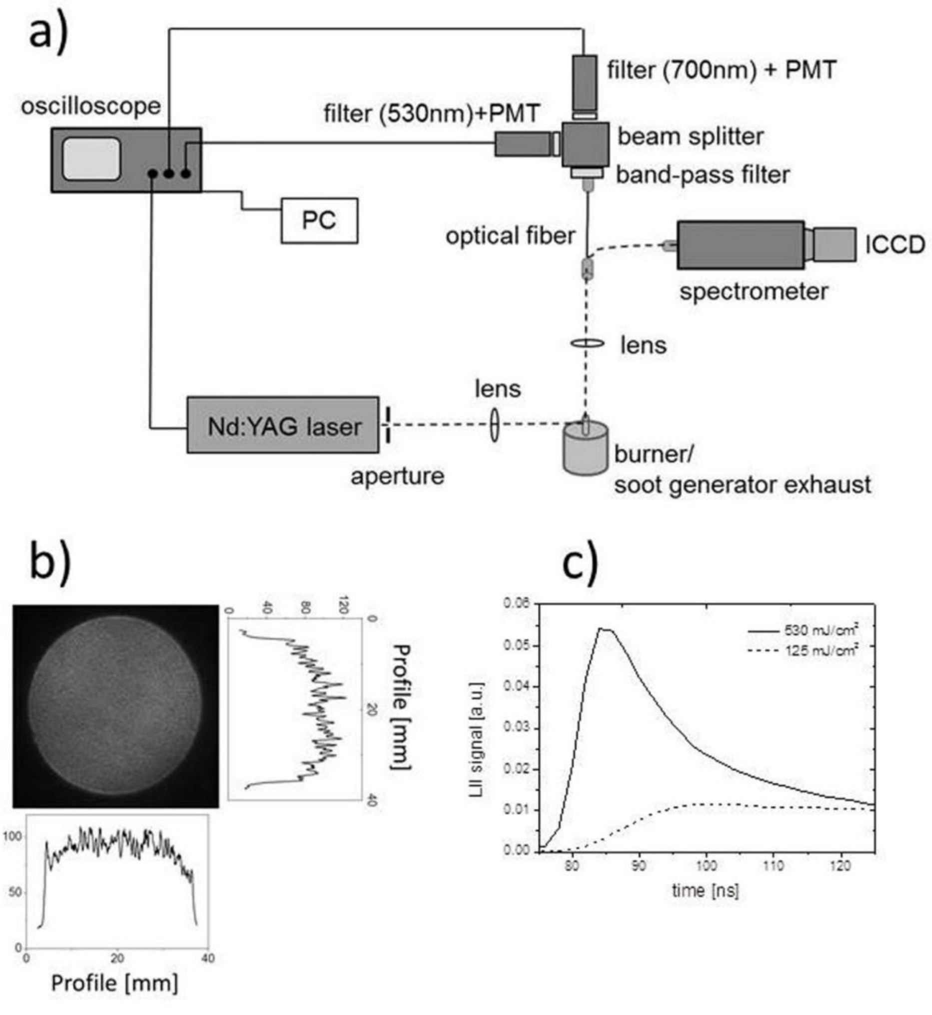

Fig. 1 a) Experimental setup for two-color and spectral LII. b) Image of the 3.5-mm laser beam and relative cross sections. c) Typical LII signalscollected at 530 nm, at low and high laser fluence.

The incandescence signal was imaged onto a 1 mm-diameter optical fiber. The other end of the fiber is properly coupled with a system of optical blocks (Hamamatsu) containing the receiving optics and the detectors. The radiation passes through a short-wavelength pass filter (CVI, $\lambda < 850$ nm) and a dichroic mirror (660 nm) to be divided into two arms where bandpass filters and PMT modules (Hamamatsu H5783-20, 078 ns rise time) were located. In order to measure LII signals in two different spectral regions, filters at 530 nm (40 nm FWHM) and 700 nm (60 nm FWHM) were used, respectively. A fast digital oscilloscope (Agilent Technologies, MS06104A, 1GHz, 4 GSa/s), triggered by the laser Q-switch, was used for data acquisition and storage. Each incandescence signal resulted from an average over 256 individual curves and was calibrated by using a calibrated tungsten ribbon lamp. The prompt signal was evaluated by taking the integral over 4 ns gate around the peak [4].

Laser-induced incandescence spectral measurements have also been performed by coupling the same optical fiber used for the two-color analysis with a Czerny-Turner spectrograph (Shamrock 303i) coupled with an ICCD (iStar 321T, Andor Technology) camera. In order to investigate the spectral feature of the blackbody emission at the maximum of the incandescence temperature, the acquisition was properly triggered and the signal was collected with a gate width of 4 ns. Spectra were collected from 300 nm up to 900 nm at low spectral resolution (150 groves /mm grating) by averaging over 500 laser shots.

Measurements were carried out in diffusion flames at atmospheric pressure as well as at the exhaust of a home-made soot generator. The burner consists of two concentric brass tubes (10 mm i.d. for fuel, 100 mm o.d. for air) and was placed on a XYZ motorized table in order to investigate the flame in different locations. Mass flow meters (Bronkhorst, AK Ruurlo, The Netherlands) were used for controlling fuels and air flow rates.

The soot generator consists of a quenched diffusion flame (9 mm i.d.), usually fed with ethylene or methane in order to change the soot load. Around the flame at a variable height above the tip of the fuel tube, an annular duct with small holes is positioned, so that a Nitrogen flow is able to quench the flame. The generated soot is confined in a cylindrical chimney (30 mm i.d., 150 mm length) and the LII measurements were performed right at the exhaust of that chimney. In order to change the soot load, the fuel, the position of the quencher with respect to the flame and the Nitrogen flow rate can be changed. In this work both flames and soot generator were fuelled with 0.18 Nl/min of ethylene or 0.38 Nl/min of methane to produce 6.5 cm height flames.

Results and discussion

2.1 Ethylene

As previously mentioned, two-color LII measurements were carried out both in flames and at the exhaust of the home-made soot generator fuelled with the same gases to investigate both in-flame and "cold" (soot generator exhaust) soot particles.

In flame measurements were performed at 30, 40 and 50 mm heights above the burner. In particular, measurements at 30 mm were carried out on the soot layer in order to reduce self-absorption effects. Similarly, the flame inside the soot generator was quenched at the same heights.

The behavior of the prompt LII signals as a function of the laser fluence are reported in Figure 2 where a comparison between the LII signals collected at 530 nm from in-flame and cold soot is shown for the three heights under investigation (30 mm (a), 40 mm (b) and 50 mm (c)). The curves have been normalized to the LII signals detected in the plateau regime. In all cases, a similar trend is observed: the LII signal increases with the laser fluence and then remains constant at laser fluences higher than 300 mJ/cm^2, with a slight decrease at the highest fluence due to sublimation effects. The cold soot LII signal is shifted to higher laser fluence values than the corresponding in-flame soot signal. Same results have been observed for the signals collected at 700 nm but they are not reported here since they do not add any further information.

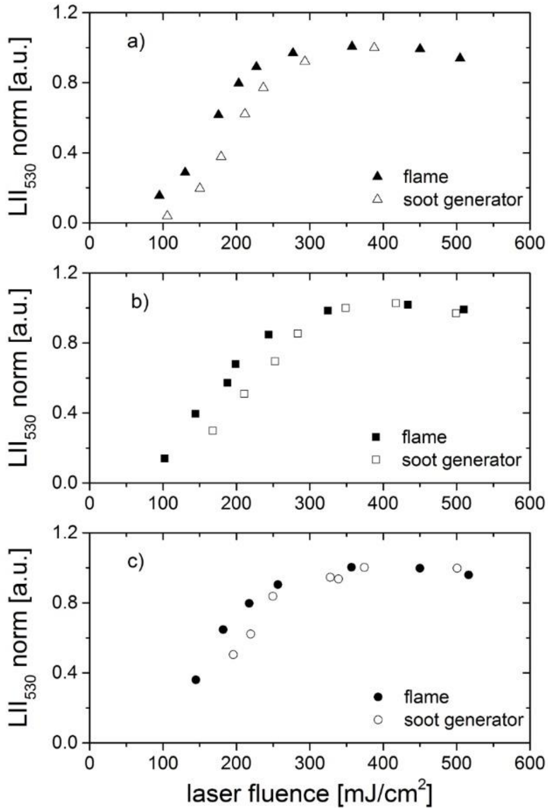

Fig. 2 Normalized LII signal collected at 530 nm versus laser fluence in flame and at the exhaust of soot generator at HAB = 30 mm (a), 40 mm (b) and 50 mm (c). Measurements refer to ethylene flame. The curves have been normalized to the LII signals detected in the plateau regime

This shift in fluence can be attributed to different effects. One effect could be a lower peak temperature reached by the cold soot in the low fluence regine according to the basic relationship [31]:

$$T_s = T_g + \frac{6\pi E(m) R_0}{\lambda_{exc}\rho_{soot}c_{soot}} \qquad (1)$$

where T_g is the gas temperature, ρ_{soot} is the soot density, c_{soot} is the soot heat capacity, R_0 is the laser fluence, λ_{exc} is the excitation laser wavelength and E(m) the soot absorption function. From Eq. (1), once the gas/particle temperature and the laser fluence are known and the value of the absorption function considered fixed, the peak of the incandescence temperature is strictly related to the initial particle temperature. Consequently, as Liu et al. also reported in their work [28], if the initial temperature decreases of 1000 °C, a shift of the incandescence temperature towards higher laser fluences (more than 50 mJ/cm^2) should be expected.

The ratio of the prompt signals collected at the two wavelengths, being related to the incandescence temperature, was analyzed and reported in Fig. 3 for both the flame (3a) and the soot generator (3b). In Fig. 3a measurements collected at different heights in flame are shown. The curves are quite overlapped and the ratio increases with the laser fluence and then remains almost constant for fluences higher than 300 mJ/cm^2. These measurements indicate that the optical properties of the in-flame soot are very similar and are in agreement with the data reported in the literature and obtained in the same experimental conditions [9]. A similar trend has been observed for the cold soot case as reported in Fig. 3b. Such behavior suggests that the incandescence ratio, and therefore the soot temperature, is independent on the quenching conditions, which in turn means on both soot concentration and, possibly, soot optical properties. However by directly comparing Fig. 3a and 3b we observe that the saturation curves for the in-flame and cold soot are quite overlapped at high laser fluences, and only a slight difference can be observed in the low fluence regime. The error of the signals ratio is about 7% and the error in the laser fluence measurements in the order of 5%. Although a clear statement can not be made about the similarity of the two curves, Fig. 3 seems to indicate that similar temperatures are reached by the soot in the flame and at the exhaust of the soot generator.

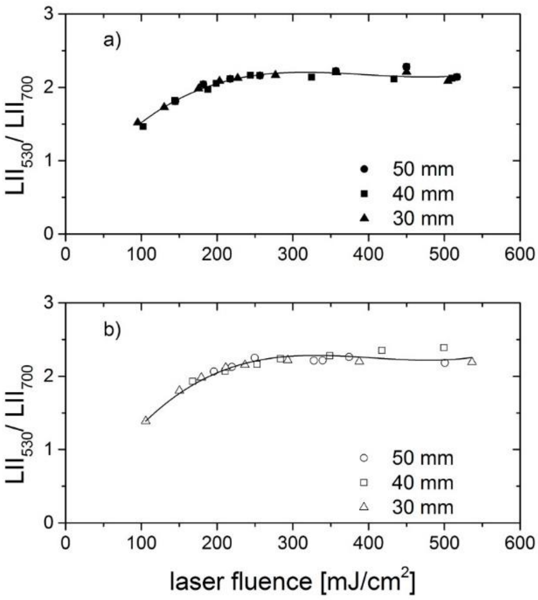

Fig.3 Prompt LII signals ratio versus laser fluence at different heights in flame (a) and at the exhaust of soot generator (b) fuelled with ethylene. In both figures, the corresponding least mean square fit of the data is also reported

Such result is very surprising and in contrast with Eq. (1) considering the initial different temperature (about 1600-1700 °C [32] for the flame and 400 °C for the soot generator). Such inconsistency can be explained by different factors that influence the saturation behavior of the LII signals such as the presence of larger aggregates at the exhaust of soot generator than in flames or a coating of the soot particles that can affect their optical properties as observed by Bambha et al. [29]. In particular, the authors observed a delay in the onset of the LII signals and in the soot peak temperature, which has been attributed to different factors, including coating thickness, coating-induced particle morphology and optical-property changes. In addition to the influence of surface absorbers or coating, the fluence shift has also been attributed to changes in the physical and optical properties of the particles due to increasing soot maturity [13, 26, 33].

Moreover, differences in the particle density and consequently in its heat capacity could somehow affect the laser absorption process. It is also important to stress, that, due to laser heating a change of the soot particle structure and consequently of the optical properties occurs [34]. As a result, increasing the laser fluence, the values of incandescence ratio could be the same independently from the initial gas temperature. Therefore, it seems pretty clear that working at laser fluences higher than 300 mJ/cm^2 would avoid the measurements to be affected by the likely different optical properties of the particles under investigation. In fact all soot particles behave in a similar way without the possibility to distinguish among different experimental conditions.

2.2 Methane

A similar analysis was also performed using methane as fuel for both flame and soot generator. In-flame soot was investigate at 46 and 50 mm heights along the flame axis, since at heights lower than 46 mm LII signal was too small to be collected at all laser fluences. On the other hand, the cold soot was obtained by quenching the flame at 40 and 50 mm, being 46 and 50 mm too close to be sure of quenching the flame at really different locations. The behavior of the LII signal versus laser fluence was also investigated.

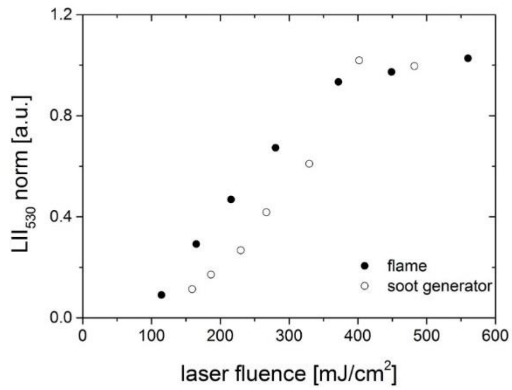

Fig. 4 Normalized LII signal collected at 530 nm in flame and at the exhaust of soot generator fuelled with methane. HAB = 50 mm. The curves have been normalized to the LII signals detected in the plateau regime.

Figure 4 shows a comparison between the LII signal collected at 530 nm for the in-flame soot at 50 mm height and the corresponding cold soot. The curves have been normalized to the LII signals detected in the plateau regime. As observed in the ethylene case, the values of the prompt LII signal of cold soot are shifted at higher fluences than the in-flame soot. This shift, of about 50 mJ/cm^2, results to be more significant than the one detected for ethylene.

The behavior of the prompt LII signals ratio versus laser fluence is reported in Fig. 5 for in-flame soot (a) and cold soot (b), respectively. The two sets of curves are quite overlapped (Fig. 5a and 5b), and all present similar trend. Interestingly, in all cases the plateau regime is reached only at 400 mJ/cm^2, that is slightly higher than the ethylene cases. All the effects previously mentioned in the ethylene case can be here considered although in the methane experiments aggregation occurs in a less significant way. It is important to emphasize the role of coating, soot maturity and other parameters strictly related to the inner structure of the primary particles [21-25]. A difference in the soot absorption function as well as in the particle density and consequently in its heat capacity could affect the laser absorption process.

A comparison between Fig. 5a and 5b shows that both curves are in good agreement in the high laser fluence regime where hardly reach a plateau value. In the low fluence regime, the two curves are more separated, with the cold soot being shifted at higher laser fluences. As a first analysis, such shift seems to be more significant than the one detected for ethylene.

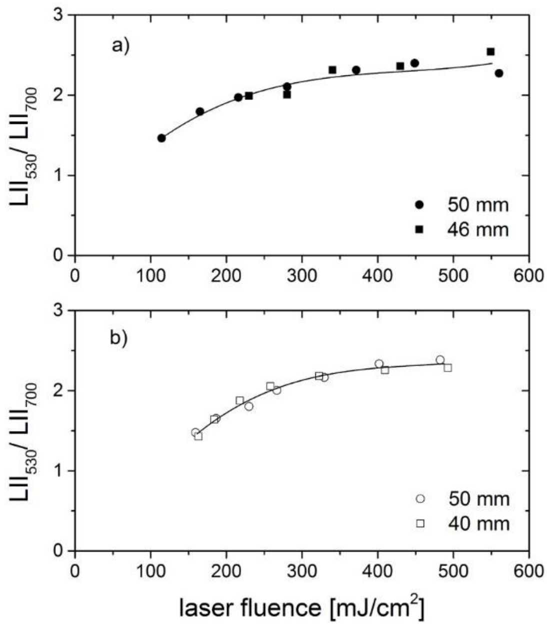

Fig. 5 Prompt LII signals ratio versus laser fluence in flame at the heights investigated (a) and at the exhaust of soot generator (b) fuelled with methane. In both figures, the corresponding least mean square fit of the data is also reported.

2.3 Comparison between ethylene and methane

For a better comprehension of the role of the fuel on soot particles absorption properties, the measurements presented in the previous sections are here rearranged in order to directly compare the ratio of the LII signals versus laser fluence for ethylene and methane. Moreover, it is also important to stress that, in this work, the change in fuel and in the experimental conditions under investigation (i.e. height in the flame, quenching height) results in a significant change in the soot load of about one order of magnitude, from 4 ppm to 0.16 ppm. In Figure 6 measurements carried out in flame (a) and at the exhaust of the soot generator (b) are shown. In all cases, for high laser fluences and in particular at values higher than about 300 mJ/cm^2(saturation curve), the curves are in good agreement. The LII signals ratio reaches a plateau value of about 2.2. The difference observed for the various experimental conditions (flame vs. soot generator, ethylene vs. methane) can be considered within the experimental uncertainties. Some differences can be observed in the low laser fluence regime. As already observed, the LII signals ratio obtained for soot particles produced by methane is shifted at higher values of laser fluences with respect to the corresponding values of ethylene. Such shift is relatively low in flames (about 30 mJ/cm^2) and increases in the measurements at the soot generator exhaust (about 50 mJ/cm^2).

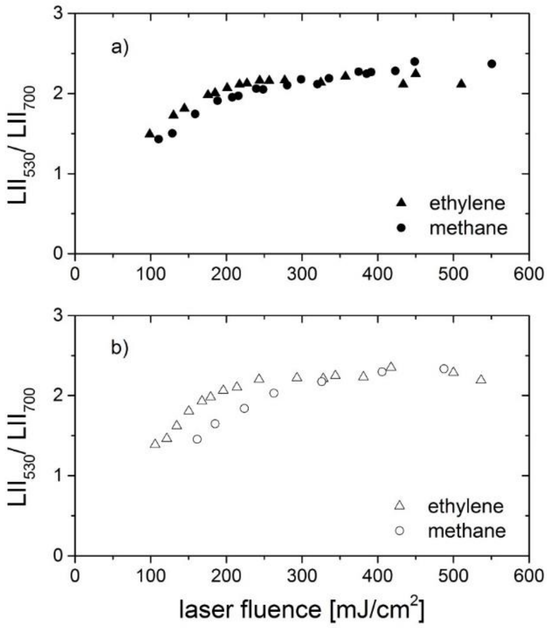

Fig. 6 Comparison of prompt LII signals ratio versus laser fluence of in-flame soot (a) and cold soot (b) obtained with ethylene and methane.

As a further analysis, to confirm the incandescence behavior in a wide spectral region, LII spectral emission measurements have also been performed. To this purpose the incandescence emission is collected at the peak of the soot incandescence temperature. Measurements have been performed at 50 mm in flames or quenching the flame inside the soot generator at the same height. In Figure 7 raw data in flame (on the left) and at the exhaust of the soot generator (on the right), normalized to the signal intensity at 700 nm, are shown. Spectra have been collected at three different laser fluences: at 500 mJ/cm^2 in the plateau regime (a, d), in the medium fluence range 210 mJ/cm^2 (b, e) and in the linear fluence regime 125 mJ/cm^2 (c, f). In order to investigate the role of the fuel, the comparison of the incandescence emission versus wavelength in ethylene and methane cases are shown, for each condition. As for the LII spectral emission at 500 mJ/cm^2, ethylene and methane exhibit the same characteristic grey-body feature, both in flame and at the exhaust of soot generator. As for the methane curve, the two small peaks observed in the in-flame soot are attributed to C_2 emission [35]. At a laser fluence of about 210 mJ/cm^2, the LII spectral emission of in-flame soot are well overlapped (Fig. 7b) while a little discrepancy is observed for the cold methane soot. In this last case the LII grey-body curves look slightly shifted toward higher wavelengths (lower temperature) in agreement with the saturation curves reported in Fig. 6b. At even lower laser fluences (around 125 mJ/cm^2) this effect looks more significant. As for in-flame soot (Fig. 7c) a very little shift of the methane spectrum toward higher wavelengths is observed compared to the ethylene one. On the other hand, for the cold soot that shift is definitively more pronounced.

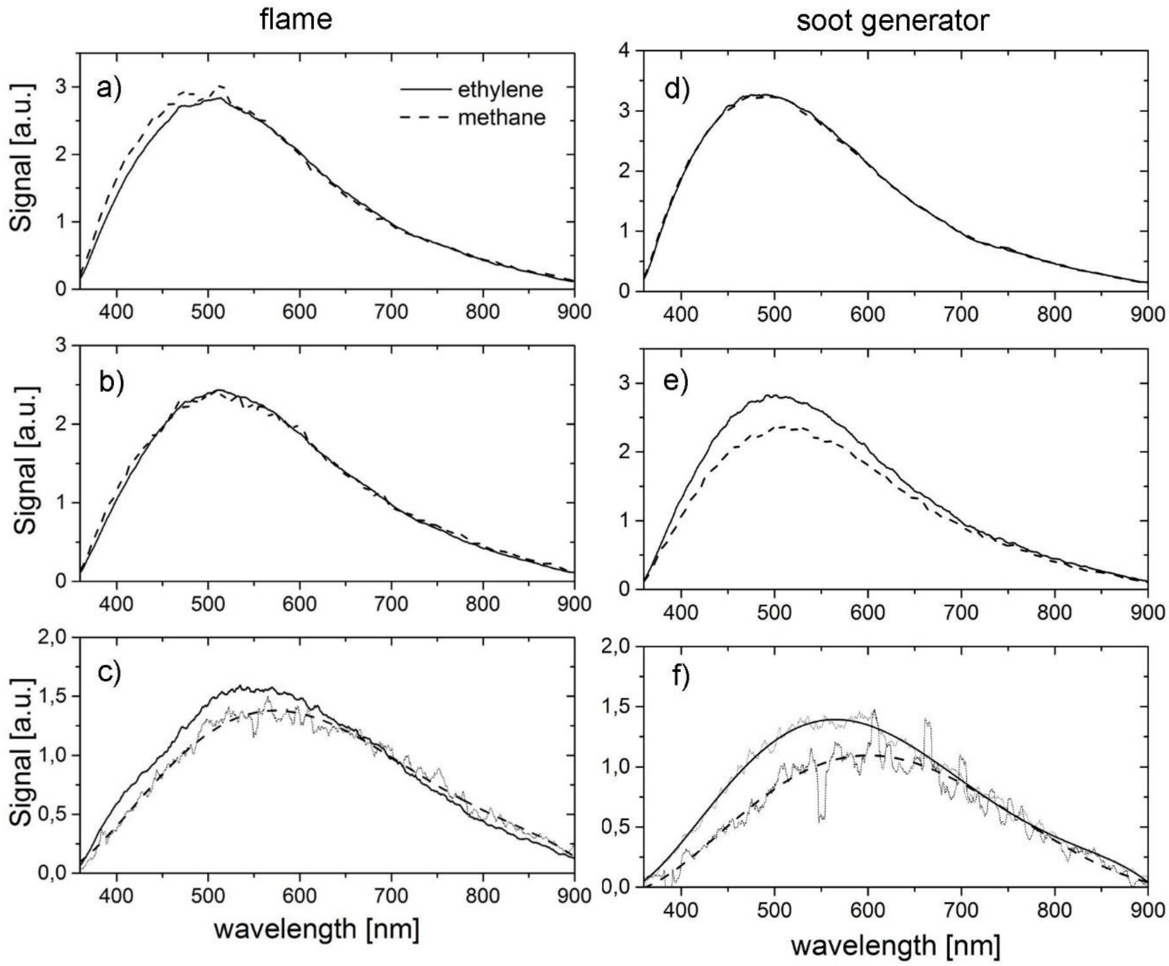

Fig. 7 LII spectral emission from flame (on the left) and from cold soot (on the right) obtained with both fuels. All spectra are normalized to the emission intensity at 700 nm. Measurements have been performed at three values of laser fluence: flame (a) and soot generator (d) at 500 mJ/cm^2; flame (b) and soot generator (e) at 200 mJ/cm^2; flame (c) and soot generator (f) at 125 mJ/cm^2.

The scenario resulting from Fig. 7 is consistent with the measurements of the incandescence signals ratio versus laser fluence previously reported. Comparing ethylene and methane flames, a slight shift of the emission intensity towards lower temperature is detected at a fixed laser fluence, that is more significant in the cold condition than in flame. The results presented in this work allow us to make some consideration about the absorption properties of soot particles in the conditions under analysis. In general, the data reported in the literature on the effect of the fuel type suggest that the difference, if any, can be considered within the measurements uncertainties [15, 16, 36]. In particular in the work of Krishnan et al., different gaseous and liquid fuels have been tested, although methane was not considered. In the work of Williams et al [36], the investigation was performed in a co-annular burner fuelled with ethylene and methane and showed no significant difference in E(m) both in the visible and in the IR spectral region. These results are not fully in agreement with the measurements reported in the present work. The shift in the laser fluence of the incandescence signal ratio, and consequently of the soot incandescence temperature of the methane data with respect to the ethylene ones, has to be attributed to some differences in the optical properties and other non-optical soot parameters, which anyway strictly affect the previous ones. Moreover, this consideration is also in line with the main structural and morphological features of the soot nanoparticles as produced in methane and ethylene flames, as it has been observed from TEM or SEM analysis [21, 22]. From such analysis, in fact, apart from the difference in the total soot load and aggregation, some changes in the internal structure, in the particle contour and the presence of coating can be observed. Finally the slight differences detected in flames become more significant considering the measurements at the exhaust of the soot generator. The lower gas/particle initial temperature could influence aggregation, coating thickness, soot maturity, which in turn affect the optical absorption properties of soot particles. More work is needed to address the relative contributions of all these effects.

Considering the LII signals calibration, soot incandescence temperature has also been calculated. To this purpose the wavelength dependent refractive index values by Chang and Charalampopoulos [20] have been used. and the derived absorption function is assumed to be constant for all the experimental conditions. Figure 8 shows the incandescence temperature behavior versus laser fluence in the different experimental conditions. At high laser fluence, an incandescence temperature of about 4100 K is reached. In the low laser fluence regime, due to the non linear relationship between the two parameters the differences in the temperature curves are less pronounced than in the incandescence ratio (see Fig. 6). However, the curve of cold soot from methane still shows an appreciable shift at higher laser fluence. The same behavior is obtained also using the soot absorption function proposed by Krishnan et al., here not reported [15, 16]. Although the different dependence of this parameter with wavelength, the overall effect of changing E(m) is an increase of the incandescence temperature of about 500°K. From the measurements and the absolute calibration the values of the soot volume fraction, f_v, are also calculated for each exepriemental condition and varying the laser fluence [4].

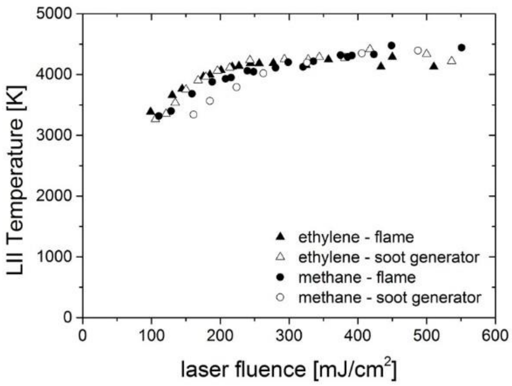

Fig. 8 Incandescence temperature behavior versus laser fluence

As an example, in Fig. 9 soot volume fraction is reported for measurements performed in the ethylene flame (a) and at the exhaust of soot generator fuelled with methane (b), respectively, at different laser fluences. A significant dependence of f_v versus laser fluence is observed in both cases.

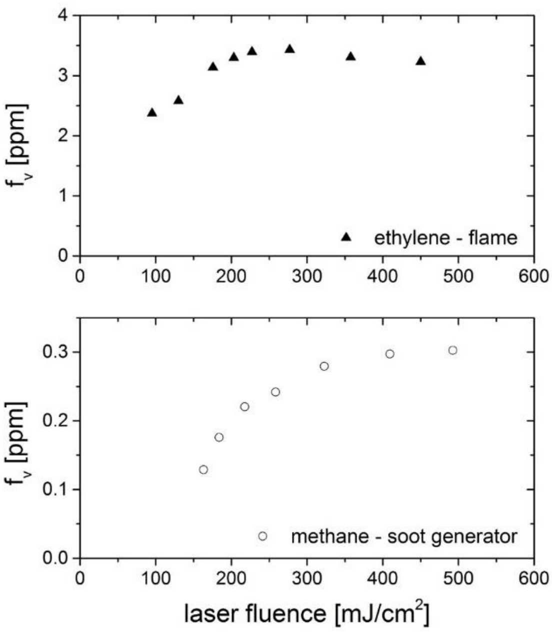

Fig.9 Soot volume fraction measurements performed in an ethylene flame (a) and at the exhaust of soot generator fuelled with methane (b), at different laser fluences.

Soot volume fraction increases with the laser fluence and tends to flatten at high values. This behavior is quite surprising, since the same value of the soot volume fraction should be obtained independently on the laser fluence used and, to our knowledge, was never observed before. This finding is not due to the particular choice of the absorption function. The application of LII measurements at low laser fluence was proposed as a better choice to reduce the perturbation of the soot particle under strong laser irradiation [8]. Unfortunately, at low laser fluence a different value of f_v is obtained compared to the ones at high laser fluences, with a change of about 30% in the worst case. These discrepancies can not be explained with shot-to-shot laser beam variations, neither with the non-uniformity of the incandescence temperature within the probe volume. Such behavior of f_v with laser fluence suggests that a change in the soot absorption parameters occurs with the increase in temperature due to laser irradiation, and in particular that the optical and non optical soot properties change due to the interaction of the laser beam with the particles.

In order to better understand the overall results reported above, the data in Fig. 9 have been further analyzed, taking for granted that the f_v value is fixed for each condition and assuming correct the value at high laser fluence. Due to f_v dependence on the absorption function through incandescence temperature and signal, some changes in E(m) with the fluence have to be considered to obtain constant f_v at all laser fluences, To this purpose the E(m) value at 530 nm (from [20]) has been kept constant, while the absorption function at 700 nm have been re-calculated at each laser fluence. The soot incandescence temperature has been then re-calculated accordingly. In Fig. 10 the $E(m)_{700}/E(m)_{530}$ ratio versus soot incandescence temperature for the two conditions is reported.

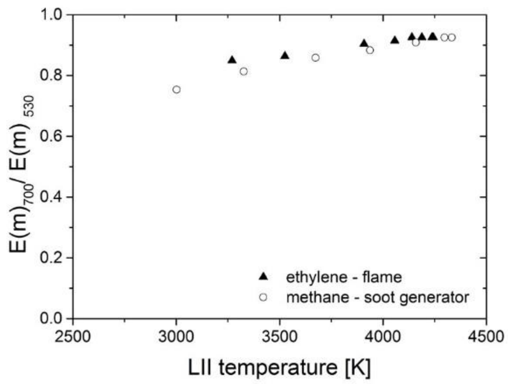

Fig. 10 Soot absorption function ratio versus incandescence temperature.

While at high temperature almost constant ratio is obtained, a decrease is observed moving towards low temperature. This behavior confirms that the soot absorption function depends on temperature. This variation in the absorption functions ratio results of a change in the temperature behavior with laser fluence, as reported in Fig. 11. In this figure the shift of the incandescence temperature towards higher laser fluences detected in the soot generator compared to flame data is more significant if compared with the corresponding trends reported in Fig. 8. The points in the low fluence regime indicate an intercept on the y axis consistent with the initial temperature of soot (400 C and 1800 C) in agreement with Eq. (1). The behavior of incandescence temperature evaluated in the other conditions under analysis is consistent with such interpretation.

As a conclusion, working at high laser fluences, the particles have been modified by the laser beam to the point that no further modification in the optical and non optical properties can occur. Therefore, working in the high laser fluence regime is more appropriate for retrieving absolute soot volume fraction values from LII measurements. This implies, however, the knowledge of the soot absorption function near the sublimation threshold.

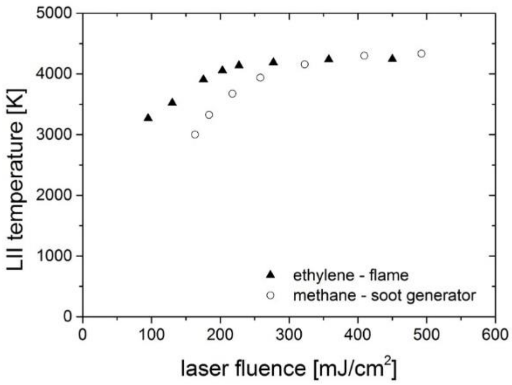

Fig. 11 Re-calculated incandescence temperature versus laser fluence.

3. Conclusions

In this work two-color laser induced incandescence measurements have been performed in atmospheric diffusion flames and at the exhaust of a home-made soot generator, both fuelled with methane and ethylene. Measurements have been carried out at several heights in flame and/or quenching the flame inside the soot generator in different positions. This allows us to investigate the influence of the fuel, the soot load, and the gas/particle initial temperature on the characteristic optical and not optical parameters of soot particles in different aging stages. Prompt LII signal at 530 nm, the ratio of the incandescence signals at two wavelengths and the incandescence temperature, as well as the soot volume fraction have been analyzed as a function of laser fluence. Moreover, LII spectral emission has also been investigated at the maximum incandescence temperature. The main results can be summarized as follows.

- The prompt LII signal from cold soot is shifted towards higher laser fluence with respect to the corresponding signal from in-flame soot.
- At high laser fluences (> 400 mJ/cm^2) the ratio of incandescence signals in all conditions is almost constant. The small discrepancies observed are within the experimental uncertainties.
- At low laser fluences some differences in the ratio of incandescence signals are observed.

In particular, a significant shift (50 mJ/cm^2) towards higher laser fluence has been obtained from incandescence measurements at the exhaust of the soot generator fuelled with methane.

- Similar observations can be made for the incandescence temperature. However, due to the non linear relationship between the incandescence signals ratio and temperature, the discrepancies at low laser fluences are less appreciable.
- The spectral feature of the incandescence signal confirms the results of the two-color LII analysis.
- Soot volume fraction have been evaluated varying the laser fluence. An increasing trend of the volume fraction has been obtained for all experimental conditions under study. In particular, a significant change of about 30% has been observed comparing the results at high and low laser fluences.
- By fixing the f_v value at all fluences, a dependence of the soot absorption functions ratio with the resulting incandescence temperature has been obtained.

The differences observed in the low laser fluence regime can be due to competitive phenomena, related to both optical and non-optical properties of the particles under analysis as well as to the initial gas temperature. Aggregation, gas/particle initial temperature, coating, aging are some of the parameters which can affect the heating processes of particles during laser irradiation. Such phenomena also affect the value of the soot volume fraction, which changes with the laser fluence. Therefore the understanding of the LII response to these properties is mandatory for a correct interpretation of the LII measurements both in the low and high laser fluence regime. More work is needed to address such issue.

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Captions

Figure 1: Experimental set-up for two-color and spectral LII.

Figure 2: Normalized LII signal collected at 530 nm versus laser fluence in flame and at the exhaust of soot generator at HAB = 30 mm (a), 40 mm (b) and 50 mm (c). Measurements refer to ethylene flame. The curves have been normalized to the LII signals detected in the plateau regime.

Figure 3: Prompt LII signals ratio versus laser fluence at different heights in flame (a) and at the exhaust of soot generator (b) fuelled with ethylene.

Figure 4: Normalized LII signal collected at 530 nm in flame and at the exhaust of soot generator fuelled with methane. HAB = 50 mm. The curves have been normalized to the LII signals detected in the plateau regime.

Figure 5: Prompt LII signals ratio versus laser fluence in flame at the heights investigated (a) and at the exhaust of soot generator (b) fuelled with methane.

Figure 6 : Comparison of prompt LII signals ratio versus laser fluence of in-flame soot (a) and cold soot (b) obtained with ethylene and methane.

Figure 7: LII spectral emission from flame (on the left) and from cold soot (on the right) obtained with both fuels. All spectra are normalized to the emission intensity at 700 nm. Measurements have been performed at three values of laser fluence: flame (a) and soot generator (d) at 500 mJ/cm^2; flame (b) and soot generator (e) at 200 mJ/cm^2; flame (c) and soot generator (f) at 125 mJ/cm^2.

Figure 8: Incandescence temperature behavior versus laser fluence.

Figure 9: Soot volume fraction measurements performed in an ethylene flame (a) and at the exhaust of soot generator fuelled with methane (b), at different laser fluences.

Figure 10: Soot absorption function ratio versus incandescence temperature.

Figure 11: Re-calculated incandescence temperature versus laser fluence.