**Propagation of ultra-short laser light pulses within spray environments**

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**Abstract**

Conventional laser diagnostics for spray characterization are limited in dense spray regions due to errors introduced by multiple scattering. The improvement and development of new laser based techniques for optically dense sprays requires suppressing the multiply scattered component from the detected signal. Due to the fact that singly scattered photons travel, on average, a shorter distance than the multiply scattered photons, a promising solution consists in temporally selecting the first portion of the signal reaching the detection area. At present, ultra-fast time-gated detection can experimentally be performed using femtosecond laser pulses (~100fs) and ultra-fast detection devices (~2ps). However, the optimization of such time-gated techniques requires predictions related to the broadening of the incident ultra-short pulse as it propagates through the spray.

In this paper, we numerically calculate photon times-of-flight within a homogeneous collection of fuel droplets by means of a Monte Carlo model specifically designed for spray diagnostics. The droplet distribution considered is typical for fuel sprays and is deduced from the modified Rosin-Rammler formula (droplet mean diameter $D = 14 \mu m$, and SMD = 23 $\mu m$). From this distribution, the averaged Lorentz-Mie scattering phase function is deduced and employed in the simulations. The scattering medium assumed is a 10 mm cubic volume and temporally resolved calculations are carried out for the back, side and forward scattering detection geometries. A large detection acceptance angle, $\theta_a = 15^\circ$, and high optical depths, $OD = 5$ and $OD = 10$, are assumed in this work. The contribution of each scattering order within the total time-resolved signal is also shown and analyzed.

For the forward scattering detection and at $OD = 10$, ~50% of the unwanted diffuse photons are removed from the detected signal when applying a 2ps time gate. However, at $OD = 5$, the amount of suppressed diffuse photons drops to ~20%. In contrast to the forward scattering pulses, back and side scattering pulses have long durations (characteristic to the dimension of the probed scattering medium) and the scattering orders are well separated in time. It is deduced, that performance in removing the contribution of diffuse photons using an ultra-fast time gate increases for smaller droplets, larger scattering volumes and higher optical depths.

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Introduction

A variety of laser based diagnostics have been developed over more than three decades in order to determine the physical properties of fuel droplets from atomizing sprays [1]. These optical measurements are of fundamental importance in the improvement of the combustion efficiency and in the reduction of pollutant emissions from modern internal combustion engines and gas turbines. However, the major limitation of the current existing laser techniques for droplet characterization is commonly related to attenuation and multiple scattering of the source radiation. Under optically thick conditions, such phenomena can introduce substantial errors in the measurements of size and concentration. Despite the development of new laser techniques recently proposed (e.g. interferometric laser imaging [2], x-ray absorption [3] or double extinction technique [4]), satisfactory quantitative and qualitative measurements are still not available due to the large contribution of multiply scattered light. One of the most promising solutions for the detection of the singly scattered light consists in discriminating the diffuse photons (which travel in average a larger distance within the spray) by temporally gating the recorded signal. This idea is supported by the availability of commercial lasers to provide ultra-short pulses (~100 fs) and by the possibility of reaching a time gate of 2 ps duration using Optical Kerr Effect (OKE) cells [5]. Furthermore, a two-dimensional time-gated shadowgraphy technique (ballistic imaging) has recently demonstrated the capability to image the dense spray region of a diesel spray [6] and of a spray in cross-flow [7] with high spatial resolution (~20 µm).

At present, the efficiency of such a modern technique to remove the diffuse light from time-gate is unknown and requires numerical calculations of the photons times-of-flight. In this article, the propagation of ultra-short laser pulses through various spray environments is investigated by means of Monte Carlo (MC) simulation. The MC model employed here has been previously applied to other studies of laser spray diagnostics [8] and has been validated against experimental results [9].

In the first section, the terminology of the relevant optical properties to describe photon transport within sprays is given. The second section is devoted to the main steps of the MC technique. A description of the simulation is provided in the third section. Results and discussion are given in section four, for the respective cases of forward, side and back scattering detection. Effects in applying a typical 2 ps gate time detection are demonstrated. Several suggestions and conclusions for the optimization of the single scattering detection from ultra-fast temporally-resolved measurements are provided in the last section.

1- Radiative transfer and photon transport in sprays

The magnitude of error introduced by multiple scattering varies with position, in a manner dependent on the source, detector, and medium geometries. Understanding effects due to multiple scattering requires knowledge of the radiative transfer of optical energy through the medium. In the field of laser diagnostics the migration of photons is generally described by the radiative transfer equation (RTE), specifying a balance of energy between the incident, outgoing, absorbing, and scattering radiation propagated through the medium. The RTE is given as:

\[
\frac{1}{c} \frac{\partial I(\vec{r}, \vec{s}, t)}{\partial t} = - \mu_a I(\vec{r}, \vec{s}, t) + \int \int \int \mu_s I(\vec{r}, \vec{s}, t) f(\vec{s}', \vec{s}) I(\vec{r}, \vec{s}', t) d\Omega d\vec{s}'
\]

where \(t\) is time, \(\vec{r}\) is the position vector, \(\vec{s}\) is the incident direction of propagation, \(f(\vec{s}', \vec{s})\) is the scattering phase function derived from the appropriate scattering theory (e.g. Lorentz-Mie theory in the case of spherical droplets), \(d\Omega\) is the solid angle spanning \(\vec{s}'\) and \(c\) is the speed of the light in the surrounding medium. The RTE can be summarized as follows: the change of radiation along a line of sight (Eq. 1 term (a)), corresponds to the loss of radiation due to the extinction of incident light (Eq. 1 term (b)) plus the amount of radiation that is scattered from all other directions \(\vec{s}'\) into the incident direction \(\vec{s}\) (Eq. 1 term (c)). The total extinction represented by Eq. 1 term (c) equals the extinction that is scattered from all other directions \(\vec{s}'\) at each light-droplet interaction.

The RTE is applicable for a wide range of turbid media; however the analytical solutions are only available in rather simple circumstances where assumptions and simplifications are introduced to reduce the equation to a more tractable form. Since there are no analytical solutions available to the transport equation in realistic cases of optical spray diagnostics, numerical techniques based on the statistical MC technique have been recently developed and utilized [1,8].

MC simulations are primarily employed to solve radiative problems within the intermediate scattering regimes where no approximation applies. Depending on the optical thickness, the scattering of light within a turbid medium can be classified into 3 regimes:

- In the single scattering regime the average number of scattering events is smaller or equal to 1 and the
non-scattered ballistic photons are dominant. For off-axis detection, the single scattering approximation which assumes that photons have experienced only one scattering event prior arriving to the detector applies.

- The intermediate single-to-multiple scattering regime operates when the average number of scattering events is between 2 and 9. In this regime, one dominant scattering order is clearly defined. No approximation can be made under such a regime.

- The multiple scattering regime is defined when the average number of scattering events is greater or equal to 10. In this regime, the relative amount of each scattering order tends to be equal and no dominant scattering order is apparent. The diffusion approximation can be applied in this regime.

<table>
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<tr>
<th>Single scattering regime</th>
<th>Intermediate scattering regime</th>
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<td>$OD \leq 1$</td>
<td>$2 \leq OD \leq 9$</td>
<td>$OD \geq 10$</td>
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Table 1. Classification of the scattering regimes as a function of the optical depth. Laser spray diagnostics operate mostly in the intermediate and in the early multiple scattering regime for diesel sprays.

By definition, the distance of light propagation between two scattering or absorbing events corresponds to the free path length $l_{fp}$. The mean free path length, which is the average distance between two light-particle interactions, is inversely proportional to the extinction coefficient, such that: $l_{fp} = 1/\mu_e$ where $\mu_e = \mu_s + \mu_a$.

Here, $\mu_s$ is the scattering coefficient and $\mu_a$ is the absorption coefficient. The optical depth ($OD$) can be calculated by dividing the total length, $l$, traversed by a light beam, by the mean free path length: $OD = l/l_{fp} = l \cdot \mu_e$. The optical depth provides an estimation of the average number of times that photons have interacted with the scattering particles, prior exiting a medium of length $l$. The classification of each scattering regime can be deduced from the value of the optical depth as presented in Table 1.

When photons propagate within turbid media, they can be categorized into various classes depending on the number of interactions they have experienced. At scattering order 0, no interaction occurs, and photons cross the spray keeping their initial direction. This group of photons is termed “ballistic”. Scattering order 1 (or single scattering) is, in optical measurements of spray, the order of interest as the information extracted from it is straightforward and directly related to the droplet characteristics (e.g. droplets size, concentration). The average photon path length through a spray is increased when larger scattering orders are considered. The range of scattering order from 2 up to 9 is associated with a group termed “snake” photons (when assuming a forward scattering detection). These photons travel a longer distance through the spray than the ballistic photons and exit the spray along approximately the same axis as the input light with a somewhat larger solid angle. Photons scattered more than 10 times are termed diffuse photons and exit the medium with a large solid angle after traveling the greatest distance in the spray.

Figure 1. Trajectories of ballistic, snake and diffuse photons crossing a collection of droplets. An illustration of the transmitted pulse is also provided.

In terms of time gating detection, the ballistic, snake and diffuse photons group, are defined as a function of their time-of-flight and time of arrival to the detector. An illustration of the three categories is given in Fig. 1. Using MC modeling, the time of flight of individual photons can be deduced from the distance traveled when exiting a spray environment. By launching a large number of photons, the pulse of light exiting the probed scattering volume can deduced with the contribution of each scattering order.

2 - The Monte Carlo technique

The implementation of MC simulation for the propagation of light radiation through turbid media has been treated by a large body of literature. Accurate descriptions of the technique can be found in [10, 11]. The fundamental steps of the MC simulation employed here are as follows: Photons enter the simulated scattering medium from an initial position with an incident direc-
The extinction coefficient is deduced such that: $\mu_e = N \sigma_e$, where $N$ is the number density and $\sigma_e$ is the extinction cross section of the scattering particles. At each particle interaction, photons can be either absorbed or scattered. If the particles are non-absorbing, the extinction coefficient is then equal to the scattering coefficient and the albedo $A = \mu_s / (\mu_s + \mu_a)$ is equal to one. In the MC technique, independent scattering is assumed requiring a distance between individual particles greater than three times the radius of the particles [12]. The MC model treats light as a collection of distinct entities, and as a consequence, interference phenomena are neglected in the simulation. This requires a random distribution of the scattering particles and the absence of periodic structures within the turbid medium. After a scattering event, the photon’s new direction is selected based on a random number and the Cumulative Probability Density Function (CPDF) calculated from the appropriate scattering phase function $f$. The scattering phase function is defined as a function of the properties of the scattering particles. Lorentz-Mie [12-13], Rayleigh-Gans [12-13] or Heney-Greenstein [14] phase functions are typically employed. The polar scattering angle $\theta_i$ defined between 0 and $\pi$ is calculated from the inverse CPDF of $f$ by: $\theta_i = \text{CPDF}^{-1}(\xi)$, where $\xi$ is once again a random number generated between 0 and 1. The resolution of the sampled scattering angle $\theta_i$ equals 0.1°. The azimuthal scattering angle $\varphi_i$ is uniformly distributed between 0 and 2$\pi$ such as the scattered radiation is assumed to be independent to the orientation of the scattering particle with respect to the direction of the incident radiation (valid for spherical particles). When a new direction of propagation is defined, the position of the next scattering point is calculated again and the process is repeated until the photon is either absorbed or exits the medium at a boundary.

The optimum number of photons employed in the simulation depends on the desired accuracy and on the characteristics of the detection. The final direction of propagation, the final position, the number of scatters, and the total path length are calculated for each light entity. If the conditions of detection are met (e.g. photon lies within the field of view of the detector with its trajectory within the acceptance angle), these data are written to disk. The process is repeated for a sufficiently large amount of photons such that the distribution of the light intensity impinging on the detector is accurately represented.

3 - Description of the simulation

The droplets are contained in a homogeneous single cubic cell of dimension $L = 10$ mm. A cylindrical flat laser beam $S$ of 30 mm diameter enters through the scattering sample crossing perpendicularly the $Y=0$ plane (back face) and exiting through the $Y=L$ plane (front face) as illustrated in Fig. 2.

![Figure 2. Illustration of the simulation; the scattering medium is a single homogeneous cube of $L = 10$ mm. The source $S$ is a cylindrical laser beam characterized by a flat spatial light intensity distribution.](image)

The source wavelength is assumed to be monochromatic such that $\lambda = 800$ nm and the light is unpolarized in the simulation. A homogeneous distribution of fuel droplets is considered from the modified Rosin-Rammler formula [15]; the probability $Q(D)$ to have a particle of diameter smaller than $D$ is given as:

$$Q(D) = 1 - e^{-D/D_k}$$

where $D_k$ is the particle diameter such that 63% of the total liquid volume is in droplets of smaller diameter and $q$ is a constant. Here, $D_k$ is fixed to 15 $\mu$m and $q$ equals 5. The mean diameter obtained equals 14 $\mu$m and the SMD equals 23 $\mu$m. The resultant PDF of the droplet distribution is given in Fig. 3. The droplets are spherical and non-absorbing with refractive index $1.4 + 0.0i$ (typical of fuel droplets). Scattering and extinction coefficients are then equal and the simulations are run with $\mu_{ext}$ fixed to 1.0 and 0.5 mm$^{-1}$. The resulting optical depths are respectively 5 and 10, corresponding to the intermediate and early multiple scattering regime. The surrounding medium is air (refractive index equals 1+0.0i). The range of particle size is 1 $\mu$m – 40 $\mu$m (typical of droplet sizes in fuel sprays) and the resulting size parameter $x = D \pi / \lambda$ is: $3.93 \leq x \leq 157.08$. 


The averaged scattering phase function \( \tilde{f} \) is calculated for the particle distributions described in Fig.3 and is illustrated in Fig.4. The resultant scattering CPDF of \( \tilde{f} \) , representative of the complete droplets distribution, is deduced in Fig.5 and used for the MC simulation. Computed photons are recorded at the exit position, provided detection conditions are met. These conditions specify that the angle between the vector normal to the detection face (front face, side face or back face) and the vector direction of the photons must be within the acceptance angle, \( \theta_a \) (see Fig. 2). The detection acceptance angle is set here to \( \theta_a = 15^\circ \).

\[
\tilde{f}(\theta_s) = \frac{\int_{0}^{\infty} n(D) \sigma_{\text{ext}}(D) f(D, \theta_s) dD}{\int_{0}^{\infty} n(D) \sigma_{\text{ext}}(D) dD}
\]

with \( \int_{4\pi} \tilde{f}(\theta_s) d\Omega(\theta_s) = 1 \) (3)

Figure 3. Droplet size distribution assumed. Here the droplets mean diameter equal 14 \( \mu \)m and the SMD equals 23 \( \mu \)m.

Droplets are assumed to be perfectly spherical such that the Lorentz-Mie theory applies [12, 13]. The local scattering phase function (within polydisperse homogeneous media) can be assumed constant and equal to the average phase function \( f \) over the complete distribution of drops size such that:

\[
\tilde{f}(\theta_s) = \frac{\int_{0}^{\infty} n(D) \sigma_{\text{ext}}(D) f(D, \theta_s) dD}{\int_{0}^{\infty} n(D) \sigma_{\text{ext}}(D) dD}
\]

The incident light pulse is considered as a Dirac pulse (infinitesimally small). The photon time-of-flight is deduced for each detected photons from their total path length within the scattering volume. Time resolved calculations are performed for the forward, side and back scattering direction respectively with a resolution of \( \Delta t = 10 \) fs. For each simulation, 2 billion photons are sent through the scattering medium and the resultant computational time is \( \sim 16 \) hours at \( OD = 10 \), when using a modern Intel(R) Core(TM) 2 CPU 6600 @ 2.40GHz processor. The relative speed of computation is then on the order of 10 \( \mu \)s/photon (at \( OD = 10 \)).

4 - Results and discussion

The first set of results shows photon arrival times for the forward scattering detection. At large optical depth \( OD = 10 \), most of the image information is contained in 13% of the overall signal level, as shown in Fig.6(a). This portion of the signal contains photons which experienced almost no deflection from their initial direction, even if, they have participated in a succession of scattering events. From the graph given in
within the combustion chamber. It shows that 17% of the light intensity detected for the case of removing the diffuse light is, in this case, not as good as wanted photons are still detected. The efficiency of wanted photons suppressed is 10% and 41% of unwanted photons over the total signal increase significantly with time. By detecting with a 2 ps time gate, the amount of unwanted photons increases. The characteristic is mostly true for the low scattering orders. In the case of the diffuse photons (tenth scattering order), the early high pick of light intensity (observed at low scattering orders) does not occur, and the light intensity decreases smoothly with time. By detecting with a 2 ps time gate it can be seen that 43.5% of the signal, corresponding mainly to the diffuse photons is suppressed. However, another 43.5% of somewhat less desirable snake photons are still detected. From these considerations, it is deduced that 50% of unwanted photons are removed from the detected signal for an optical depth set to 10, assuming a detection acceptance angle $\theta_a = 15^\circ$. When the optical depth is reduced to $OD = 5$, the amount of desired photons over the total signal increase significantly and reaches 49% of the total signal. By time gating the signal with a 2 ps time gate, the amount of unwanted photons suppressed is 10% and 41% of unwanted photons are still detected. The efficiency of removing the diffuse light is, in this case, not as good as for the case of $OD = 10$, and equals 20%. The calculation shows that 17% of the light intensity detected within $\Delta t = 10$ fs, corresponds to the ballistic photons. Within this same time interval, expected results demonstrating: $I_1 > I_2 > I_3 > I_4 > I_5$ are shown. The contribution of diffuse photons is very low at $OD = 5$ and remains fairly constant in time.

The dimension of the scattering volume plays a crucial role in the efficiency of removing the diffuse photons contribution. In the investigated simulations, the scattering medium is a cube of 10 mm sides. Assuming a scattering volume with dimension twice as big with equal optical properties (same $OD$ and distribution of droplets) the distance traveled by the photons would be twice as long, effectively doubling the pulse duration. Therefore, a given time gated device can work well for large dimensions of scattering volume even at relatively low optical depth.

The second set of results (Fig.7) shows photon arrival times for side scattering detection. In contrast to the case of forward scattering detection, duration of the detected pulse is much longer (around 90 ps – which is related to the dimension of the scattering volume) and does not change when varying the optical depth (similar shape and duration). Furthermore, the maximum intensity related to each scattering order does not occur at the same time, but is shifted from one scattering order to the next (the maximum amount of singly scattered light intensity occurs before the maximum amount of secondly scattered light intensity which occurs before the maximum amount of thirdly scattered light intensity and etc). This time separation is reduced when increasing $OD$. It is also observed that the dominant scattering order and the value of the optical depth remain equal for both cases. Below this value, the light intensity increases with scattering order and above this value the light intensity decreases with scattering order.

The third set of results (Fig.8) shows the photon arrival times for back scattering detection. A smooth decay of the light intensity with time is observed. This decay is more important at the high optical depths. A second decay is observed in Fig.8(c), at $t = 70$ ps. This decay shows the limit of the scattering medium for which the backscatter signal drop to zero. However, at $OD = 10$ this limit is no longer present (due to the increase of photon path lengths $\geq 2L$). The maximum light intensity detected corresponds to photons which have been singly backscattered at the entrance of the scattering medium. The contribution of this maximum reduces abruptly, whereas the contribution from higher scattering orders increases in time until reaching a maximum value. Each scattering order becomes dominant in turn.

**Conclusion**

Time resolved calculations of photon time-of-flight within spray environments have been investigated by Monte Carlo modeling. Results for the forward, side, and back scattering detection have revealed the following features:

- For the forward scattering detection: The scattering orders are not separated at the very early detection times due to the very large forward scattering lobe exhibited by the fuel droplets (~15 $\mu$m). A 2ps time gate removes 50% of diffuse photons at $OD = 10$ and 20% at $OD = 5$. Better efficiency is expected for more isotropic scattering particles (smaller droplets), scattering media of larger dimension and at larger optical depths.

- For the side scattering detection: The extraction of the singly scattered light is not efficient at optical depths larger than 5. However, at lower optical depth, a 2 ps time gate would be particularly efficient if applied when the amount of single scattering reaches its maximum value.

- For the back scattering detection: Information related to the optical depth, to the droplets size and to the dimension of the spray probed can be deduced from the decay of the back scattered light.
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References
Figure 6. Laser light intensity of an ultra-short laser pulse transmitted through a spray environment (cubic volume of $L = 10$ mm side) versus time. In (a) - (b) $OD = 10$ and in (c) - (d) $OD = 5$. 
Figure 7. Laser light intensity of an ultra-short laser pulse detected on the side face of a spray scattering medium (cubic volume of $L = 10$ mm side) versus time. In (a) - (b) $OD=10$ and in (c) - (d) $OD = 5$. 
Figure 8. Laser light intensity of an ultra-short laser pulse detected on the back face of a spray scattering medium (cubic volume of $L = 10$ mm side) versus time. In (a) - (b) $OD=10$ and in (c) - (d) $OD = 5$. 