# Temporal scattering of dense scattering media under ultra short laser light illumination: Application for particle sizing.

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#### Abstract

We intend to demonstrate the contribution of ultra short laser light methods for improving the particle sizing of dense scattering medium like plumes or engine sprays. We especially focus our attention on the time-resolved propagation of ultra short laser pulses in the forward and backward directions considering a Monte-Carlo (MC) scheme. We investigate the influence of the detector-source configurations on the retrieval of spectral backscattering and extinction coefficients derived from the measurements of the spectral transmittance and backscattering power. An efficient method developed to retrieve Particle Size Distribution of aerosols from multi-spectral LIDAR returns is then adapted to the investigated case. This method is finally tested on the numerical spectral backscattering and extinction coefficients derived from MC simulations.

#### **1** Introduction and Context

For aerosols particle sizing, we developed a specific scheme inverting spectral extinction and backscattering coefficients that could be derived from single scattering multispectral LIDAR returns using Klett method [1]. This inverse scheme includes a self-regularization method built from the data uncertainties.

Assuming an homogeneous scattering medium of length L, the extinction  $\alpha(\lambda)$  and backscattering  $\beta(\lambda)$  spectral coefficients could be retrieved from respectively the measurement of spectral transmittance  $T(\lambda) \propto \exp(-\alpha(\lambda) \times L)$  or backscattering power  $P(\lambda) \propto \beta(\lambda) \times T^2(\lambda)$ .

In LIDAR application, retrieving extinction  $\alpha(\lambda)$  and backscattering  $\beta(\lambda)$  spectral coefficients is only performed from backscattering power  $P(\lambda)$  and consequently, requires to presume a known relation between  $\alpha(\lambda)$  and  $\beta(\lambda)$ . Using simultaneously  $P(\lambda)$  and  $T(\lambda)$  prevents from this requirement.

Nevertheless, the characterization of the particle size distribution (PSD) in dense scattering media like plumes, engine sprays... using a spectral measurement of the transmission and the backscattering power is a challenge as multiple scattering phenomenon scrambles useful information.

Retrieving the Particle Size Distribution f(R) (PSD) from these spectral data requires:

- a reduction of the contribution of multiple scattering events and consequently of the errors occurring on the derived spectral coefficients. Time-resolved analysis of ultra short laser pulses propagation in the backward and forward directions seems to indicate a temporal separation of ballistic and scattering photons. As a consequence, studying such phenomenon could be used in order to reduce multiple scattering if it is related to appropriate detector configurations (FOV, time gate...).
- an efficient inverse method unaffected by the residual uncertainties. The method developed for LIDAR applications could be adapted for this purpose.

In the first paragraph, we present the Monte-Carlo (MC) scheme developed to simulate ultra short laser pulses propagation in dense scattering media.

### 2 Monte-Carlo Simulations

The MC scheme developed is based on a temporal photon pursuing method [2] that we have extended to include polarization according to the Stokes formalism. To increase the convergence of the MC results, a pseudo MC approximation is used to evaluate the temporal intensities collected by two small detectors geometries in the forward and backward directions [3].

The statistical scattering properties are derived from the temporal Mueller matrices  $P_{ii}(\theta,t)$  or the

integrated ones  $\overline{P}_{ij}(\theta) = \int_{\Delta t} P_{ij}(\theta, t) dt$ . Those matrices are evaluated through a scanning of frequency

coupled with a Lorenz-Mie (LM) theory algorithm associated with a FFT transform of the incident pulse and an inverse FFT transform of the scattering matrices [4]. The Figure 1 presents the temporal phase function of  $a50\mu m$  sphere illuminated by a 50fs pulse for different times relative to the transmitted time of flight. We can observe that the angular direction, where occurs the maximum of light scattered, is time dependant.

The Figure 2 compares the integrated linear depolarization ratio to this obtained for the same sphere and a continuous illumination. The pulse integrated ratio smoothes the fluctuations of the "continuous".



Figure 1: Temporal phase function normalized to maximum  $P_{11}(\theta, t) / \max(P_{11}(\theta, t)) \forall \theta$ (Sphere radius = 50µm and 50fs Pulse)



Figure 2: Ratio  $\overline{P}_{12}(\theta)/\overline{P}_{11}(\theta)$  for  $\Delta t = 2500$  fs and a 50 fs pulse width compared to the same ratio obtained for a continuous wave.

We here specially focused our attention on scattering medium, in which large optical thicknesses (OT), various bimodal Particle Size Distributions (PSD) and concentration gradients could be observed. The spectral transmittances  $T(\lambda)$  or backscattering received powers  $P(\lambda)$  of those media could be then derived considering various detector-source geometries, temporal pulse widths and integration ranges The Figure 3 represents an example of the transmitted temporal intensities considering various collection angles.



Figure 3: Transmitted temporal intensities considering various collection angles (.the arrow indicates an increase of the collection angle)

#### 2 Inverse scheme

The spectral transmittances  $T(\lambda)$  and the backscattering received powers  $P(\lambda)$  are affected by multiple scattering, which depends on the optical thickness and the chosen experimental configuration. The first challenge is then to derive an optimal experimental configuration (time gate, spectral range, detector FOV, pulse width...) where the temporal integration of transmitted and backscattered temporal intensities leads to measured intensities that are close to those related respectively to a collimated or a single scattering measurement hypothesis.

Nevertheless, the spectral extinction  $\alpha(\lambda)$  and backscattering  $\beta(\lambda)$  coefficients derived from the previous relations could be still uncertain. As a consequence, retrieving the Particle Size Distribution f(R) (PSD) from these spectral coefficients requires regularization.

We propose to use an inverse scheme developed for LIDAR applications. It includes a self-regularization method built from the uncertainties  $\Delta X(\lambda_i)$  associated to a set of mean data values  $X(\lambda_i)$ . This method is based on previous works [5] derived for angular scattering data and spectral extinctions [6]. It is adapted to consider simultaneously spectral extinction and backscattering coefficients. This method is actually numerically tested to retrieve the PSD of atmospheric aerosols from extinction  $\alpha(\lambda)$  and backscattering  $\beta(\lambda)$  spectral coefficients. These coefficients are obtained from Klett inversion [1] of noisy

multispectral LIDAR backscattering signals. The Figure 4 presents the PSD of a maritime bimodal aerosol retrieved from an "8 wavelengths (UV to NIR)" Simulated LIDAR Signals (SLS). A 10% Gaussian noise is added to the theoretical extinction  $\alpha(\lambda)$  and backscattering  $\beta(\lambda)$  to perform SLS. The retrieved PSD are derived considering one set of spectral data (extinction or backscattering) or both of them. For this case, we can observe that only considering backscattering data is insufficient and lead to a worth estimation of the PSD.



Figure 4: Retrieved PSD compared to theoretical one.

## **3** Further works

An optimal experimental configuration (time gate, spectral range, detector FOV, pulse width...) reducing multiple scattering contributions for the transmittance  $T(\lambda)$  or backscattering received powers  $P(\lambda)$  and having a sufficient Signal to Noise Ratio (SNR) is first evaluated. The extinction and backscattering coefficients retrieved from the inversions of the Beer-Lambert law and the single scattering LIDAR return equation are then compared to the theoretical values used for the MC simulations in order to evaluate the residual uncertainties due to multiple scattering.

In parallel and considering various spectral data issued from diverse scattering media, we also try to evaluate the influence of the spectral range and the mesh grid on the PSD recovering method. The influence of the uncertainties level on the inverse scheme efficiency is also estimated and related to the field of application of the proposed method.

Finally, a complete simulation is performed for a specific scattering media and the retrieved PSD are compared to the theoretical ones used in the MC simulation; in order to assess the proposed concept.

These numerical results will be presented in the poster session.

A time gated experimental setup based on a femtosecond laser associated to a Non-colinear Optical Parametric Amplifier (NOPA) is also under development. Preliminary experimental results could be presented if they are available.

#### References

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