Non-linear optical response of aerosol nanoparticles exposed to femtosecond laser light

Doctorant·e

JORET MAXIME

Direction de thèse

YON Jerome (Directeur·trice de thèse)
MAZUR MAREK (Co-encadrant·e de thèse)

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Rapporteurs de la thèse

DEVEL MICHEL SUPMICROTECH-ENSMM
OUF FRANCOIS XAVIER Laboratoire national de métrologie et d'essais (LNE)

Membres du jurys

BARVIAU BENOIT, , URN - Université de Rouen Normandie
DEVEL MICHEL, , SUPMICROTECH-ENSMM
DUPRAY VALÉRIE, , URN - Université de Rouen Normandie
GEHIN EVELYNE, , Université Paris-Est Créteil
MAZUR MAREK, , INSA Rouen Normandie
OUF FRANCOIS XAVIER, , Laboratoire national de métrologie et d'essais (LNE)
PETITPREZ DENIS, , Université de Lille
YON Jerome, , INSA Rouen Normandie

Abstract

Aerosols, fine particles suspended in the air, significantly impact climate change by affecting the Earth's radiative balance. They also have health implications when deposited in the lungs. Additionally, aerosols play a role in various processes, such as black carbon production for tires, incorporation into materials to modify their properties, and thermal radiation in combustion processes. Controlling the concentration, size, and morphology of these particles is crucial due to their desired or undesired effects. Optical diagnostics are essential for in situ assessment of these properties, offering high spatial and temporal resolutions. Static Light Scattering and Dynamic Light Scattering are elastic processes widely used to determine particle size, volume, and concentration. Induced or natural emission techniques are also employed to assess volume fraction and temperatures. However, conventional optical diagnostics cannot provide information on the particles' internal structure (e.g., graphitization state, composition) or their specific surface area (surface of aerosol divided by their mass). These quantities are of high interest, particularly for evaluating toxicological impacts, which strongly depend on particle morphology. Recent advancements in intense and short laser pulses have enabled the generation of nonlinear optical (NLO) effects, including Second Harmonic Generation (SHG), Third Harmonic Generation (THG), and Hyper-Rayleigh Scattering (HRS). These effects have shown significant potential in biosciences and fundamental physics for investigating surface-sensitive phenomena. While some experiments have been conducted on particles in solution or nanoparticles on droplet surfaces, SHG by nanoparticles in the aerosol phase has not yet been demonstrated. The objective of this thesis is to address this gap and evaluate the potential of NLO for the metrology of fine particles, such as soot particles produced during the incomplete combustion of fuels. Specifically, this work aims to explore SHG, THG, and HRS by black carbon particles and other types of nanoparticles to assess new measurands. The experiments involved developing a dedicated and innovative optical setup capable of analyzing the NLO response resolved in time, wavelength, and angle. This setup can isolate SHG, THG, and HRS from other NLO phenomena, such as laser filamentation. The optical setup was calibrated to quantify the generated signal and optimized for high sensitivity while avoiding NLO generation from its own optical elements. The results confirm that soot particles, DEHS droplets, and arc-generated carbon nanoparticles exhibit SHG/ HRS at intensities more than seven orders of magnitude smaller than static light scattering. THG, if present, was not detectable and is so even weaker. SHG and HRS depend on aggregate and monomer size but do not seem to be influenced by the organic or elemental carbon content. The experiments also show that SHG-HRS increases linearly with particle surface area, independent of particle shape and composition. Furthermore, the angular response differs fundamentally from static light scattering, exhibiting an isotropic nature. This suggests that aerosols are prone to generating Hyper-Rayleigh Scattering, which is found to be promising to quantify the specific surface area of an aerosol in situ. By providing access to this crucial information, the present work paves the way for a new class of laser-based diagnostics for aerosols.