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Multiple light scattering diagnosis of combustion products

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ABSTRACT

Accurate measurements of microstructural properties of solid propellants are crucial for evaluating the performance of propulsion systems. Besides, the particulate size and number density determine an increase of the heat flux to the combustor walls. Supervision of particulate emission in propulsion burners is required for understanding the basic physical mechanism and for monitoring the temporal evolution of combustion process. Accordingly, in situ characterization of particulates is highly desired and, due to their noninvasive and remote characteristics, optical techniques are a suitable candidate. Knowledge of radiative properties, extinction and scattering of solid particles is of prime interest in understanding the optical characteristics of the propellant medium and of the combustion products [1]. The optical cross-sections of the particles are determined by their size, density and chemical composition and are commonly evaluated, for homogeneous spheres, by means of the Mie theory [2]. In addition, the characterization procedures are complicated by the fact that large agglomerates with complex morphologies are generated in the burning process. Due to substantial optical depths and a large particle number density, optical waves propagate through multiple scattering processes. For the estimation of number of densities and particle size distributions of burning products, we study the efficiency of scattering-based transmission and reflection methods. Scattering regimes are identified for typical situations and appropriate continuous-wave scattering techniques are discussed in the context of characterizing the particulates. In a burner, waves propagation is also affected by thermal-induced refractive index fluctuations. However, the time scales of these fluctuations are much larger than the time scales involved in wave propagation through the burner and, therefore, only scattering and absorption of solid particulates essentially affects a pulsed measurement. The effectiveness of time-resolved transmission and reflection techniques is also discussed.

INTRODUCTION

Electromagnetic wave scattering in random media is a challenging, yet to be solved problem with broad interdisciplinary interest. Applications range from microbiology to materials science to atmospheric propagation. Solid propulsion systems are also an area where knowledge of physical properties of particles such as sizes and concentration is desired in order to optimize/control the burning mechanism. The burning propellants as well as the combustion products are described as discrete particles media where scatterers occupy random positions. Due to high scattering contrast and long paths, optical wave propagation though particulate media is dominated by multiple scattering. A rigorous scattering solution is certainly impossible but, however, reasonable approximations can lead to practical methods to retrieve particle size and shape, spatial distribution, and clustering.

The intensity and polarization of light scattered by particulate media depend on shape, surface roughness, absorption, and internal scattering coefficient of the particles. The inversion procedures, however, are complicated by the fact that the photons suffer multiple scattering inside the composite medium and only averaged quantities can be revealed by an optical measurement. Recent advances in the theory of multiple light scattering showed that, in spite of an overall randomness of the scattered field, there are also steady-state, coherent phenomena such as enhanced backscattering (EBS) or time-resolved scattering (TRS) that allow structural information retrieval [3-6].

EBS PHENOMENON

Until recently, the coherent light propagating through random media has been considered to be somehow degraded, losing its coherence properties. When coherent light is scattered by a random medium, interference effects between the scattered waves, which have traveled through the medium along different paths, occur. The random pattern of interference is called *laser speckle*. When the individual scatterers are allowed to move over distances of the order of the wavelength or more, the distribution of intensities in the speckle pattern is averaged out and becomes essentially flat. However, one kind of interference still survives in this average, the interference of the waves emerging from the medium

in directions close to the backward direction and which have travelled along the *same* light path but in *opposite directions*. Accordingly, an enhancement of the scattered intensity develops in a narrow angular cone around the backward direction.

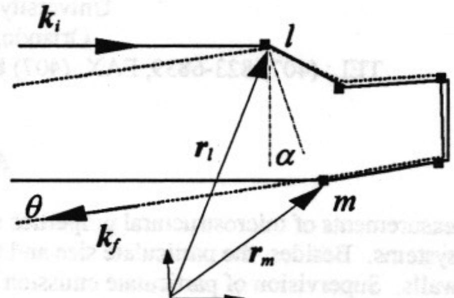


Figure 1 **Principle of enhanced backscattering phenomenon. In the exact backscattering direction (θ=0) the paths are equally long. In all other directions there will be a phase difference between the time-reversed waves.**

Let us consider the situation depicted in Fig. 1. We concentrate on two scattering centers located at r_l and r_m in the random medium and on interference between waves that have these scatters as starting and respective ending points of their tortuous paths. Waves that have $\mathbf{r}_l$ and $\mathbf{r}_m$ as ending points and that have traveled through the same scattering centers but in reversed order (time- reversed or momentum-reversed scattering sequence) will have experienced the phase difference

$$\Delta\Phi = \frac{2\pi}{\lambda}(\mathbf{k}_i - \mathbf{k}_f)\cdot(\mathbf{r}_l - \mathbf{r}_m) \tag{1}$$

where, λ is the wavelength of light and $\mathbf{k}_{i,f}$ are the initial and final wave vectors as shown in Fig. 1. In the exact backscattering direction constructive interference occurs because $\mathbf{k}_i$= -$\mathbf{k}_f$ and, therefore, $\Delta\Phi$=0 independent of the position of the scattering centers l and m. Evaluating the dot product in Eq. 1, one obtains

$$\Delta\Phi = \frac{2\pi}{\lambda}|\mathbf{r}_l - \mathbf{r}_m| cos\left(\alpha - \frac{\theta}{2}\right) sin\left(\frac{\theta}{2}\right) \propto \frac{2\pi}{\lambda}|\mathbf{r}_l - \mathbf{r}_m| cos(\alpha) sin\left(\frac{\theta}{2}\right) \tag{2}$$

In the experimental situation of small scattering angles, Eq. 2 can be approximated by

$$\Delta\Phi \approx \frac{2\pi}{\lambda}|\mathbf{r}_l - \mathbf{r}_m|\theta \tag{3}$$

Thus, the angular width of the constructive interference developed around backscattering direction (θ=0) is of the order of

$$W \approx \frac{1}{2\pi}\frac{\lambda}{D} \tag{4}$$

where D is the averaged distance (over the entire scattering volume) between scattering centers of type l and m. Accordingly, the EBS angular width is controlled by the average distance D and, therefore, reflects the properties of a scattering medium.

EBS AND PARTICLE SIZING

When optical waves propagate through particulate media with grain (particle) sizes larger than the wavelength of light, the photon path is built up by successive reflections and refractions. At each interface between voids and grains (between particles and suspending medium) part of the radiation is refracted and part is reflected. A coefficient of reflection R, averaged over all angles of incidence, can be calculated using the Fresnel coefficients

$$R = \frac{1}{\pi}\int_0^{\frac{\pi}{2}}\left[R_\perp(n,\theta) + R_\parallel(n,\theta)\right] cos(\theta) d\theta \tag{5}$$

where $n=n_r+in_i$ is the complex index of refraction of the particle. For particles with small absorption coefficients (n_i), R is usually less than 0.1 while the refraction coefficient R-1 is almost unity. In the final energy balance, a good approximation is therefore provided by considering the photon paths as being developed inside grains (particles). Accordingly, the scattering mean free path (the average distance D as it has been defined in the precedent section) equals, in average, the particle size A and the width of an EBS cone directly relates to the average particle size A

$$W \approx \frac{1}{2\pi}\frac{\lambda}{A} \tag{6}$$

The relation of inverse proportionality between the width of the coherent enhancement (a measurable quantity) and the average particle size can be used to infer particle sizes in particulate suspensions. The idea is supported by experimental measurements. A schematic of the measuring apparatus is presented in Fig. 2. The main elements are a coherent radiation source (laser) with the corresponding beam expander, a collection optics assembly, polarizing optics, and a detector that can provide a spatially resolved intensity measurement.

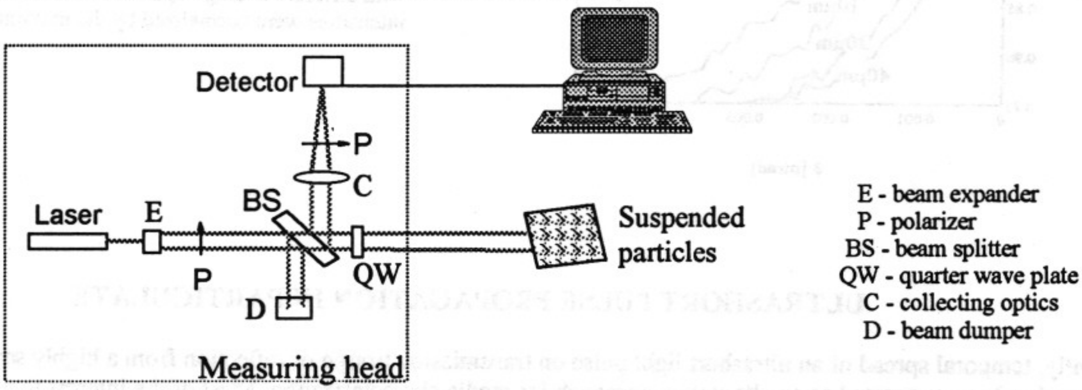
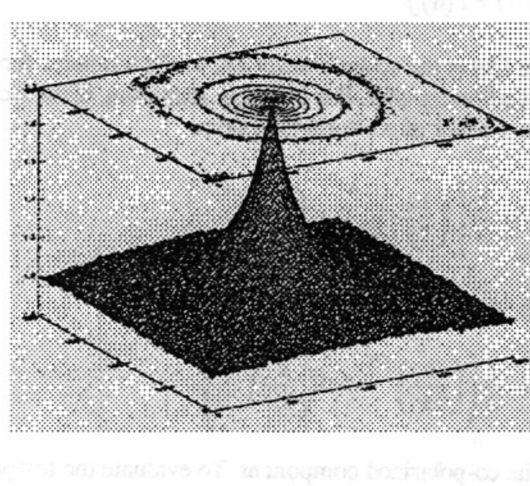


Figure 2 Principle of enhanced backscattering measuring apparatus.

The polarization optics, in the configuration shown in Fig. 2, is needed to terminate single-scattering contributions to the measured signal. These contributions could originate from both scattering volume (single-scattering from the investigated particles) as well as from scattering cell window or other optical surfaces. The spatially-resolved detector can be a linear or a two-dimensional CCD array. The electric signal provided by the detector is processed by the acquisition and analysis system, which is PC-based. In Fig. 3, we present a typical cone of enhanced backscattering recorded with the apparatus described in Fig. 2.

Figure 3 Two-dimensional image of a typical cone of enhanced intensity obtained in the exact backscattering direction.



Systematic measurements were performed on Al2O3 samples with polyhedral grains of different sizes. Because of their relative smooth surfaces, the possibility of light scattering on the grain surfaces is ruled out. In addition, the present set of samples is characterized by small size dispersion. For all the samples, a definite cone of enhanced intensity was observed and, in Fig. 4, we present the central parts of the cones corresponding to different particle sizes. In order to clearly observe the W variation with the average particle size A, the intensities were normalized by the maximum value of each angular cone. The definite dependence found between the width of the enhancement cone and the average particle size confirm our hypothesis that multiple-light-scattering-based techniques developed in a diffusive approach have strong potential as characterization tools for particulate media.

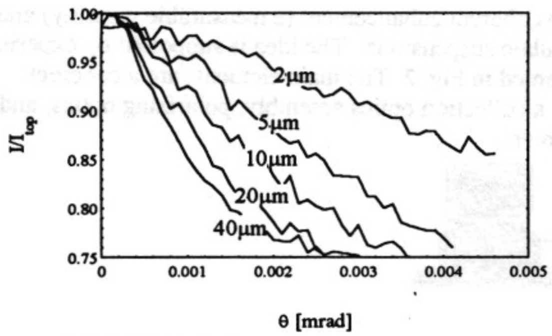


Figure 4 Co-polarized enhancement of backscattered intensities corresponding to media with different averaged particle sizes. The intensities were normalized by the maximum

ULTRASHORT PULSE PROPAGATION IN PARTICULATE

Recently, temporal spread of an ultrashort light pulse on transmission trough or reflection from a highly scattering medium has been suggested as an alternative approach for media characterization. Most of the interest focussed on obtaining useful images trough significant depth of scattering media and involved a series of time-gating techniques. An ultrashort input pulse will typically broaden by orders of magnitude depending on the overall size of the sample as well as on the intrinsic scattering and absorption properties of the particulate medium. Consequently, a time-gated measurement of transmission or reflection will reveal structural information such as average particle size and particle number density.

We investigated the temporal spread of femtosecond pulses in reflection from dense particulate media consisting of suspensions of micron-sized silica particles. The intensity profiles of backscattered pulses in the two polarization channels are described as a function of number of scattering events by

$$I^{p,s}(n) = I(n) f^{p,s} \tag{7}$$

where $I(n)$ is the scalar component of the temporal decay predicted by diffusion theory and the factors $f^{p,s}$ describe the polarization transfer. For isotropic scattering, these factors are estimated from the Bethe-Salpeter equation as a function of the number of scattering events n

$$f^{p} = \frac{1}{3}\left[\left(\frac{10}{15}\right)^{n-1} - \left(\frac{7}{15}\right)^{n-1}\right] \tag{8}$$

for the cross-polarized component and

$$f^{s} = \frac{1}{3}\left[\left(\frac{10}{15}\right)^{n-1} + 2\left(\frac{7}{15}\right)^{n-1}\right] \tag{9}$$

for the co-polarized component. To evaluate the temporal profile of the backscattered intensity

$$I(t) = \sum_{n=1}^{\infty} I(n) c(n,t) \tag{10}$$

the contribution *c(n,t)* of the *n*-the order scattering event to the total light intensity has to be known. A Poisson distribution that depends on the value of transport mean free path l^* has been suggested

$$c(n,t) = \frac{\left(\frac{ct}{l^*}\right)^n}{n!\left[\exp\left(\frac{ct}{l^*}\right) - 1\right]} \qquad (11)$$

Using a background-free cross-correlation technique with very high temporal resolution, the reflected pulse profiles in the co- and cross-parallel polarization channels were recorded for samples with different volume fractions. Typical backscattered pulses corresponding to media with various solid loads are presented in Fig. 5.

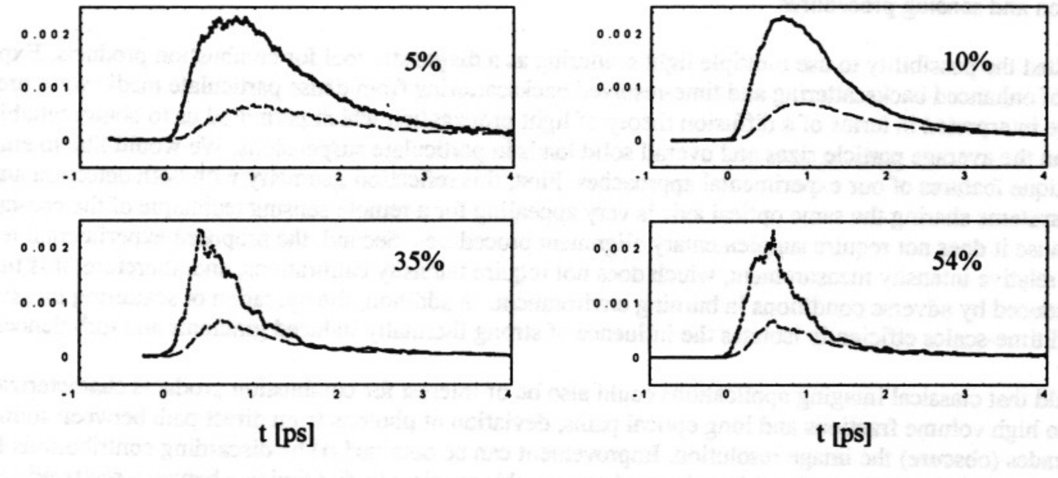


Figure 5 Backscattered light pulses from dispersions of micron-size silica particles with volume fractions as indicated. Laser pulses with duration of 150 fs were used to cross-correlate backscattered signals from the samples.

We have found that the temporal shapes of the backscattered pulses do not change significantly when increasing the volume fraction of particles and all curves could be fitted well by a functional form derived in the frame of the diffusion approximation as predicted by Eq. 7. We used the pulse profiles in the parallel-polarization channel to infer the transport mean free path l^* for samples with different volume fractions and the results are summarized in Fig. 6.

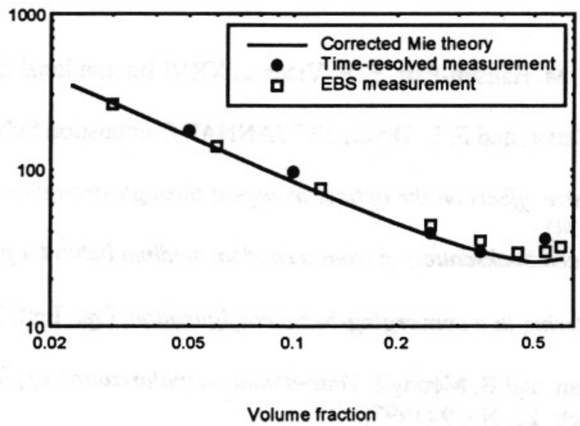


Figure 6 Values of the transport mean free path corresponding to particulate media with different volume fractions. Also shown are corresponding values inferred from enahanced backscattered measurements as well as values estimated from Mie scattering theory.

A typical multiple-scattering process is characterized by an average scattering mean free path *l*, an averaged cosine of the scattering angle *g*, and a transport corrected mean free path $l^*=l/(1-g)$. The value of *l* represents the average distance between scattering centers and *g* includes the effect of scatterer type and size. Note that there are, in principle, several ways to measure l^*: time-resolved, transmission or reflection techniques. The results presented in Fig. 6 prove that multiple light scattering techniques can be used to accurately infer a wide range of solid loads in particulate suspensions.

CONCLUSIONS

The difficulty of *in-situ* measurements at high densities combined with a tedious averaging procedure over different sites of the structure usually precludes a direct evaluation of macroscopic properties of combustion products. The alternative is to assess the random structure over larger scales by means of a scattering technique. With a long history of applications, light scattering is one of the most attractive alternatives, which, beside its intrinsic noninvasive and nonperturbative character, has also the potential for developing high-performance, noninvasive, and real-time instrumentation and sensing procedures.

We have studied the possibility to use multiple light scattering as a diagnostic tool for combustion products. Exploratory experiments of enhanced backscattering and time-resolved backscattering from dense particulate media were presented and results are interpreted in terms of a diffusion theory of light propagation which permitted us to obtain reliable information on the average particle sizes and overall solid loads in particulate suspensions. We would like to emphasize here some unique features of our experimental approaches. First, this reflection geometry with both detection and illumination systems sharing the same optical axis is very appealing for a remote sensing technique of the combustion products because it does not require supplementary alignment procedures. Second, the proposed experimental techniques are based on relative intensity measurement, which does not require intensity calibrations, and, therefore, it is insensitive to changes induced by adverse conditions in burning environment. In addition, the operation of scattering measurements at picosecond time-scales efficiently isolates the influence of strong thermally induced gradients and turbulences.

We should add that classical imaging applications could also be of interest for combustion products characterization. Of course, due to high volume fractions and long optical paths, deviation of photons from direct path between source and detector degrades (obscure) the image resolution. Improvement can be obtained (i) by discarding contributions from scattered photons and using conventional imaging techniques (this requires to discriminate between scattered and non-scattered photon by time-resolved, polarization-resolved, optical tags, coherence-gated, and spatial filtering) or (ii) by understanding and using the scattered photons and discriminate between "useful photons" and noise. It is a long time belief that no useful information can be found in the *diffuse* scattering from regions containing a large number of scattering centers. However, recent advances in understanding the multiple wave scattering phenomena suggest the possibility to retrieve structural information via diffuse light. Possible directions for further development should be based on improved understanding of multiple scattering phenomena, novel design for imaging (scattering) techniques, new photon sources and detection systems with specific time, polarization, and wavelength properties, new image assessment, enhancement, that account for quantitative aspects of multiple scattering contributions.

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