Generic formulation of a generalized Lorenz-Mie theory for pulsed laser illumination.

By

G. Gouesbet, L. Mees, and G. Gréhan

Laboratoire d'Energétique des Systèmes et Procédés,
Unité Mixte de Recherche 6614/CORIA
CNRS/INSA et Université de Rouen,
BP 08,
76 131 Mont Saint Aignan, France

ABSTRACT

During the two last decades, a vigorous effort has been devoted to the study of interactions between arbitrary shaped beams (for continuous waves) and particles. When the particle under study allows one to solve the scattering problem by relying on the separability of variables, we say that the associated theory is a generalized Lorenz-Mie theory (GLMT). At the present time, GLMTs are available for the following king of particles: homogeneous spheres, multilayered spheres, ellipsoids, infinitely long cylinders (with circular or elliptical cross-sections), sphere with one arbitrarily located spherical inclusion, and aggregates. Such a theories are relevant to the field of optical particle characterization in two-phase flows (phase-Doppler systems, rainbow refractometry, shadow Doppler techniques, for instance).

The aforementioned GLMTs are extensions of simpler theories when particles are illuminated by continuous plane waves. Another line of extension is by considering the case of illumination by pulses (single pulses or train of pulses), viewed as continuous arbitrary shaped beams modulated by a pulse envelope.

Nowadays, pulsed laser beams become of common use, with a growing interest for the generation and applications of femtosecond pulses. In particular, pulsed lasers may activate many interesting nonlinear phenomena in microcavities, such as stimulated Raman scattering, stimulated Brillouin scattering, third-order sum generation or lasing. They open the way for new optical particle characterization techniques, for instance concerning measurements of chemical species concentration in droplets.

It is therefore desirable to possess a GLMT for the case of particles illuminated by laser pulses, with arbitrary spatial supports. Such a GLMT will be presented in this paper. This GLMT is said to be generic. The word "generic" means that the theory is presented under a form allowing one to consider arbitrary scattering particles.

1. INTRODUCTION

During the two last decades, a vigorous effort has been devoted to the study of interactions between arbitrary shaped beams (for continuous waves) and particles. When the particle under study allows one to solve the scattering problem by relying on the separability of variables, we say that the associated theory is a generalized Lorenz-Mie theory (GLMT). At the present time, GLMTs are available for the following king of particles: homogeneous spheres, multilayered spheres, ellipsoids, infinitely long cylinders (with circular or elliptical cross-sections), sphere with one arbitrarily located spherical inclusion, and aggregates. Such a theories are relevant to the field of optical particle characterization in two-phase flows (phase-Doppler systems, rainbow refractometry, shadow Doppler techniques, for instance)[1, 2, 3].

The aforementioned GLMTs are extensions of simpler theories when particles are illuminated by continuous plane waves. Another line of extension is by considering the case of illumination by pulses (single pulses or train of pulses), viewed as continuous arbitrary shaped beams modulated by a pulse envelope.

Nowadays, pulsed laser beams become of common use, with a growing interest for the generation and applications of femtosecond pulses. In particular, pulsed lasers may activate many interesting nonlinear phenomena in microcavities, such as stimulated Raman scattering, stimulated Brillouin scattering, third-order sum generation or lasing. They open the way for new optical particle characterization techniques, for instance concerning measurements of chemical species concentration in droplets.

It is therefore desirable to possess a GLMT for the case of particles illuminated by laser pulses, with arbitrary spatial supports. Such a GLMT will be presented in this paper. This GLMT is said to be generic. The word "generic" means that the theory is presented under a form allowing one to consider arbitrary scattering particles.

The paper is organized as follows. Section 2 presents the general formulation and is a summary of a material developed in [4]. Also, section 3 considers the case of a Rayleigh dipole allowing one to present an analytical discussion of the formulation. Section 4 discusses the more realistic case of a sphere illuminated by a pulsed Gaussian beam (Gaussian envelope), with plane wave or Gaussian spatial supports. Section 5 is a conclusion.

2. GENERAL FORMULATION.

We consider a pulsed electromagnetic wave of the form:

$$X_i(r,t) = X_0 X_i^s(r) \exp(2ipn_0 t) g(t) \qquad (1)$$

in which X designates either E (electric field) or H (magnetic field), i is a component subscript (in a coordinate system matching the shape of the particle), r is a point in space, $X_i^s(r)$ describes the laser shape and is called the spatial support of the pulse, ν_0 is the carrier frequency, g(τ) is the (real) pulse envelope and τ is a shifted time:

$$t = t - z/c \qquad (2)$$

in which t is the time, z the axis of propagation of the beam, and c the speed of light. This formulation is quite general and is obtained by multiplying the general description of a continuous wave laser beam [5] by the pulse envelope g(τ). The pulse propagates in a transparent medium and, for convenience in this paper, g(τ) represents an unique pulse (not a train of pulses).

We assume that we are allowed to introduce a pair of Fourier transforms reading as:

$$G(n) = \int_{-\infty}^{+\infty} g(t) \exp(-2ipnt) dt \qquad (3)$$

$$g(t) = \int_{-\infty}^{+\infty} G(n) \exp(+2ipnt) dn \qquad (4)$$

where G(ν) is the pulse envelope spectrum. Conditions of existence for the pair are not discussed here, but, in general, the Fourier transform should be interpreted in terms of mathematical distributions [6, 7] (see next section). The envelope g(τ) being real, its spectrum is Hermitian:

$$G(n) = G(-n)^* \tag{5}$$

leading to:

$$g(t) = \int_0^{+\infty} G(n)\exp(+2ipnt)dn + \int_0^{+\infty} G(n)^* \exp(-2ipnt)dn \tag{6}$$

From Eqs (1) and (6):

$$X_i(r,t) = X_0 X_i^s(r)\left[\int_0^{+\infty} G(n-n_0)\exp(+2ipnt)dn + \int_0^{+\infty} G(n+n_0)^* \exp(+2ipnt)dn\right] \tag{7}$$

showing that the pulsed illumination can be decomposed into an integral of elementary monochromatic waves. The response to each monochromatic wave is assumed to be known (for instance, use a continuous wave GLMT). The response of the scatterer to an excitation $X_0 X_i^S(r)\exp(\pm 2ipnt)$ then reads as $X_i^{cw}(r,\pm n)\exp(\pm 2ipnt)$. This response of the scatterer to a pulse then is:

$$X_i^p(r,t) = \left[\int_0^{+\infty} G(n-n_0) X_i^{cw}(r,n)\exp(2ipnt)dn\right] + \left[\int_0^{+\infty} G(n+n_0)^* X_i^{cw}(r,-n)\exp(+2ipnt)dn\right] \tag{8}$$

From Hermiticity equation(5), Eq (8) becomes:

$$X_i^p(r,t) = \tilde{F}^{-1}\left[G(n-n_0) X_i^{cw}(r,n)\right] \tag{9}$$

in which $\tilde{F}^{-1}$ designates an inverse Fourier transform and the tilde means that the variable conjugated with (is the time t (not the shifted time τ).

RAYLEIGH DIPOLE

To illustrate the formulation, we first consider a case which can be analytically solved. Let us consider a Rayleigh dipole illuminated by a pulse with a plane wave spatial support, propagating along z, with the electric field oscillating parallely to x. Eq (1) becomes:

$$E_1 = E_0 \exp(2ipnt) g(t) \tag{10}$$

and we take an envelope:

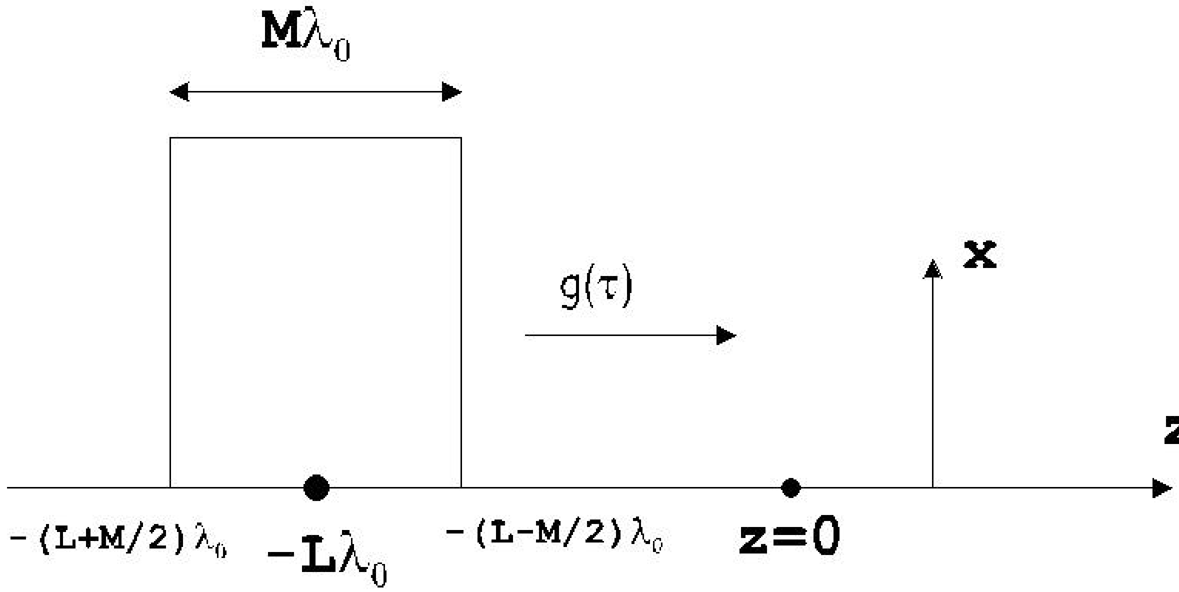

Figure 1 : Representation of the pulse envelope, at time t=0.

$$g(t) = \prod\left(\frac{t - L/n_0}{M/n_0}\right) \qquad , \text{L,M} > 0,\ \text{L>M/2} \tag{11}$$

displayed in Fig 1, for t=0. A Rayleigh dipole is located at z=0. The pulse excitation reaches the dipole at t=(L-M/2)λ_0/c and leaves it at t=(L+M/2)λ_0/c. We establish:

$$\prod\left(\frac{t - L/n_0}{M/n_0}\right) = \frac{M}{n_0}\prod\left(\frac{t}{M/n_0}\right) * d(t - L/n_0) \tag{12}$$

in which (is the Dirac distribution and (*) designates a convolution product. Using:

$$\mathbf{F}[S*T] = \mathbf{F}[S] \times \mathbf{F}[T] \tag{13}$$

in which S and T are distributions and, **F** is a direct Fourier transform, we have:

$$G(n) = \frac{M}{n_0}\frac{\sin p n M/n_0}{p n}\exp(-2ip nL/n_0) \tag{14}$$

The response of a Rayleigh dipole to an excitation $E_0 \exp(2ip n_0 t)$, for a scattered electric field oscillating along x, propagating toward positive z's, with a constant complex refractive index reads as [8]:

$$E_0 \frac{K}{z} n^2 \exp(2ip nt)\exp(2ip nz/c) \tag{15}$$

in which K is a constant.

Using Eqs (9) and (14), we afterward establish:

$$E_1^p(z,t) = E_0 \frac{KM}{zn_0}\tilde{F}^{-1}\left[n^2\right] * \tilde{F}^{-1}\left[\exp(2ip nz/c)\right] \tag{16}$$

$$* \tilde{F}^{-1}\left[\frac{\sin p(n-n_0)M/n_0}{p(n-n_0)}\exp(-2ip(n-n_0)L/n_0\right]$$

We evaluate separately each inverse Fourier transform, in particular:

$$\tilde{F}^{-1}\left[n^2\right]=-\frac{d''(t)}{4p^2} \tag{17}$$

and use:

$$d''*T=T'' \tag{18}$$

to establish:

$$E_1^p(z,t)=-\frac{K}{4p^2}\frac{E_0 n_0}{zM}\frac{d^2}{dt^2}\left[\prod\left(\frac{t-z/c-L/n_0}{M/n_0}\right)\exp(2ipn_0(t-z/c))\right] \tag{19}$$

The second derivative is evaluated with the following ingredients:

1. the derivative of a convolution product of two distributions is obtained by only deriving one distribution.

2. We have:

$$\prod(x)=H(x+1/2)-H(x-1/2) \tag{20}$$

in which **H** is the Heaviside distribution.

3. We have:

$$H'(x)=d(x) \tag{21}$$

We then obtain:

$$E_1^p(z,t)=-\frac{KE_0}{4p^2 z}\left\{-\frac{4p^2 n_0^3}{M}\prod\left(\frac{t-z/c-L/n_0}{M/n_0}\right)\exp(i2pn_0(t-z/c))\right\}+\underset{\text{Dirac singularities}}{} \tag{22}$$

The singularities (full expression not given), depending on δ and δ', occur at times when the scattered pulse reaches and leaves the location z>0. Since a Rayleigh dipole has no inertia, we then see that the causality principle is satisfied, illustrating the coherence of the formulation.

3. FINITE SPHERE

We now consider a more realistic case, requiring the use of numerical evaluations.

3.1 Configuration under study

The configuration under study is displayed in Fig 2. A pulsed laser beam, with a Gaussian spatial support characterized by its beam waist radius ω_0, its pulse time width Δt (full length at half-width) and its carrier wavelength λ_0, is scattered by a homogeneous spherical particle. The particle has a diameter d, a complex refractive index m=n-ik, and its location with respect to the beam waist center is (x_0 ,y_0 ,z_0). We aim to predict the properties of the scattered light in the direction defined by an angle (, in the far field domain).

The results are displayed as intensity versus a time delay. The time delay is defined as the time difference between the arrival time of the scattered light at the detector and the arrival time of a virtual ray which would travel, in free space, to the particle center and thereafter from the particle center to the detector. With such a definition of the time delay, we recover the classical geometrical optics formulation used in Phase Doppler anemometry [9, 10]. Optical path differences may also be evaluated as a time delay times the speed of light. As a consequence, reflected light, with an optical path smaller than the reference path, exhibits a positive time delay while, conversely, all the other scattering modes exhibit a negative time delay.

3.2 Exemplifying results, pulsed plane wave illumination.

We consider the case of a 100 µm particle diameter illuminated by a 100 fs pulse, with the center of the particle located at the beam waist center ($x_0 = y_0 = z_0 = 0$). The carrier wavelength of the illuminating beam is 0.6 µm. For convenience, we assume that the complex refractive index of the material does nor depend on the wavelength (remember that the pulse envelope generates a wavelength spectrum).

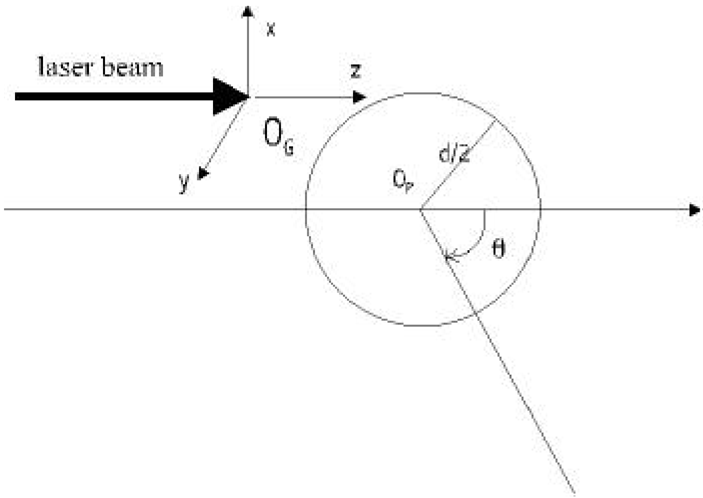

Figure 2 : The configuration under study, with the polarization defined by φ = 0

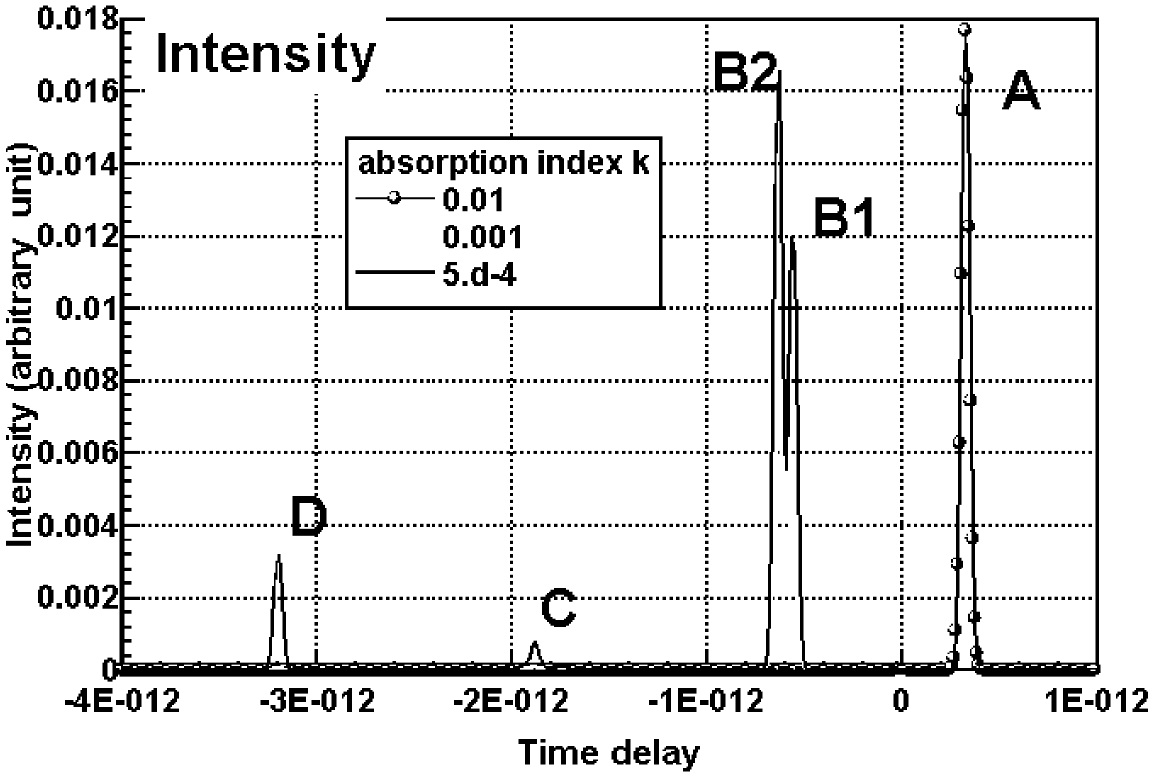

Figure 3 : Pulsed plane wave backward scattering on a water droplet

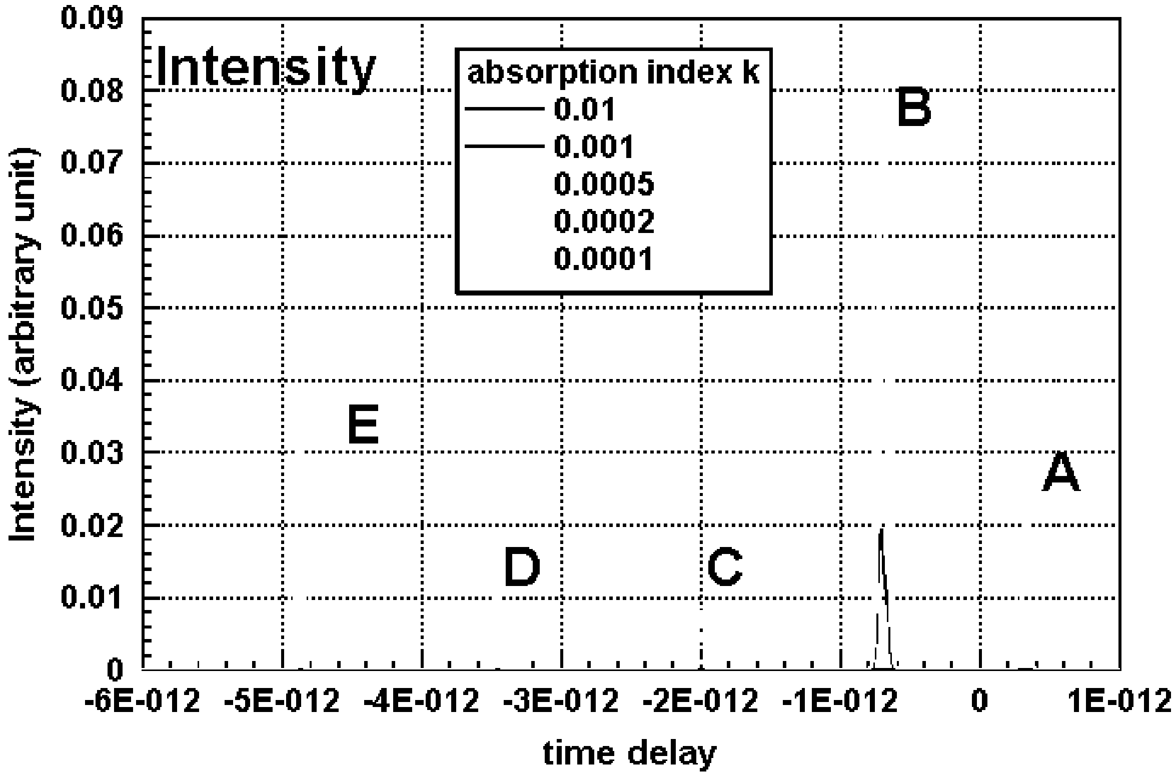

Figure 4 : Pulsed plane wave backward scattering on a glass sphere.