Measurements of Beam Shape Coefficients in Generalized Lorenz-Mie Theory and the Density-Matrix Approach. Part 2: The Density-Matrix Approach

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(Received: 4 March 1996)

Abstract

In Part 1, it was demonstrated that intensity measurements on an actual beam in the laboratory allow one to determine so-called density matrices related to the beam shape coefficients encoding the beam and used in many expressions of the generalized Lorenz-Mie theory. In this paper, expressions in the generalized Lorenz-Mie theory are rewritten and it is shown that they can be expressed in terms of the density matrices rather than in terms of beam shape coefficients, leading to what is called the density matrix approach to the generalized Lorenz-Mie theory. The possibility of rewriting the generalized Lorenz-Mie theory in terms of quantities describing the illuminating beam and experimentally measurable in the laboratory offers new opportunities in optical characterization.

1 Introduction

Generalized Lorenz-Mie theory (GLMT) describes the interaction between an arbitrary-shaped beam and a spherical particle (Ref. [1] and references therein). In this theory, the illuminating beam is encoded in a double set $\{g_{n,TM}^m, g_{n,TE}^m\}$ of beam shape coefficients (BSCs), with n ranging from 1 to infinity and m from $(-n)$ to $(+n)$. In the special case of so-called axisymmetric (on-axis) beams, such as for on-axis Gaussian beams, the double set reduces to a single set $\{g_n\}$ of special BSCs [2]. GLMT expressions related to the properties of the light scattered by the sphere are expressed in terms of these BSCs which, up to now, were evaluated by relying on a theoretical description of the illuminating beam.

In Part 1 [3], it was demonstrated that, by relying on intensity measurements on the actual beam in the laboratory, it is possible to measure density matrices related to the BSCs. In the case of axisymmetric beams, we may introduce an observable density matrix

$$I_{nm} = G_n G_m^* \tag{1}$$

in which G_ns are effective BSCs related to the BSC g_ns by

$$G_n = \sqrt{\frac{k}{\omega\mu_0}} E_0(K+i)g_n \tag{2}$$

where k is the wavenumber of the light, ω its angular frequency, μ_0 the magnetic capacity of the free space, E_0 the electric strength and K a polarization parameter which may also be measured in the laboratory.

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In the general case, when the beam is described by a double set of coefficients $g_{n,TM}^m$, $g_{n,TE}^m$, we had to introduce three observable density matrices:

$$M_{np}^{mq} = g_{n,TM}^m g_{p,TM}^{q^*} \tag{3}$$

$$E_{np}^{mq} = g_{n,TE}^m g_{p,TE}^{q^*} \tag{4}$$

$$C_{np}^{mq} = g_{n,TM}^m g_{p,TE}^{q^*}. \tag{5}$$

In the case of axisymmetric beams, when the beam with an $\exp(i\omega t)$ time dependence propagates along the axis Oz of a Cartesian coordinate system, from negative to positive z, BSCs g_n and $g_{n,TM}^m$, $g_{n,TE}^m$ are related as follows [2]:

$$\left.\begin{array}{l} g_n^m = 0, |m| \neq 1 \\ \dfrac{g_n}{2} = g_{n,TM}^1 = \dfrac{1}{K} g_{n,TM}^{-1} = i g_{n,TE}^1 = -\dfrac{i}{K} g_{n,TE}^{-1} \end{array}\right\}. \tag{6}$$

The fact that the polarization parameter K intervenes in the relations defining g_n versus $g_{n,TM}^{-1}$ or $g_{n,TE}^{-1}$ explains why it explicitly appears in the density matrix I_{nm} (Eqs. (1) and (2)) whereas it does not appear in the density matrices M_{np}^{mq}, E_{np}^{mq}, C_{np}^{mq} [3].

The observability of all these density matrices is only of interest if many significant quantities appearing in GLMT may be expressed in terms of them rather than in terms of BSCs. This is the issue addressed in this paper. The answer is positive, leading to a reformulation of GLMT that we call the density matrix approach to GLMT. The possibility of rewriting the GLMT in terms of beam quantities measurable in the laboratory offers new opportunities in optical characterization.

The paper is organized as follows. Section 2 describes the density matrix approach for axisymmetric beams. Section 3 describes the density matrix approach in the general case (arbitrary shaped beams). In an Appendix, we provide a justification of the density matrix terminology.

0934-0866/97/0204-0088 $17.00+.50/0

2 Axisymmetric Beams

GLMT expressions associated with the scattered light are mostly usually given with a normalization condition [4]:

$$\frac{1}{2}\sqrt{\frac{\epsilon_0}{\mu_0}}E_0E_0^* = \frac{1}{2}\frac{k}{\omega\mu_0}E_0E_0^* = \frac{1}{2}E_0H_0^* = 1. \tag{7}$$

From Eqs. (2) and (7), we then have a normalization relation between I_{nm} and $g_n g_m^*$:

$$g_n g_m^* = \frac{I_{nm}}{2(K^2+1)}. \tag{8}$$

Scattered properties are described in an (r, θ, φ) spherical coordinate system attached to the center of the scatterer (see Ref. [4] for details of the geometry of the problem which is not useful to repeat here, and Figure 1 in Part 1 [3]).

In the general case, scattered intensities in the far-field are expressed as [4]

$$\begin{pmatrix} I_\theta^+ \\ I_\varphi^+ \end{pmatrix} = \frac{\lambda^2}{4\pi^2 r^2}\begin{pmatrix} |S_2|^2 \\ |S_1|^2 \end{pmatrix} \tag{9}$$

in which λ is the wavelength and S_2, S_1 are generalized amplitude functions given by

$$S_2 = -ikr\frac{E_\theta}{E_0}e^{ikr} \tag{10}$$

$$S_1 = -kr\frac{E_\varphi}{E_0}e^{ikr} \tag{11}$$

where

$$E_\theta = \frac{iE_0}{kr}e^{-ikr}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n}\frac{2n+1}{n(n+1)}\left[a_n g_{n,TM}^m \tau_n^{|m|} + imb_n g_{n,TE}^m \pi_n^{|m|}\right]e^{im\varphi} \tag{12}$$

$$E_\varphi = \frac{-E_0}{kr}e^{-ikr}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n}\frac{2n+1}{n(n+1)}\left[ma_n g_{n,TM}^m \pi_n^{|m|} + ib_n g_{n,TE}^m \tau_n^{|m|}\right]e^{im\varphi} \tag{13}$$

Here, a_n and b_n are the usual scattering coefficients of the classical Lorenz-Mie theory for plane waves and $\pi_n^{|m|} = \pi_n^{|m|}(\cos\theta)$, $\tau_n^{|m|} = \tau_n^{|m|}(\cos\theta)$ are generalized Legendre functions, defined by

$$\pi_n^m(\cos\theta) = \frac{P_n^m(\cos\theta)}{\sin\theta} \tag{14}$$

$$\tau_n^m(\cos\theta) = \frac{dP_n^m(\cos\theta)}{d\theta} \tag{15}$$

where P_n^m are the associated Legendre polynomials. Eqs. (12) and (13) exemplify the fact that GLMT offers a solution to the light scattering problem in terms of three sets of parameters: (i) BSCs (that depend on the structure of the laser beam as well as on the particle location), (ii) Mie scattering coefficients (a_n and b_n) that essentially pertain to particle properties, i.e. size parameter and complex refractive index times the size parameter, and (iii) functions that depend on the observation point, in particular special angular functions that depend on the scattering direction under consideration.

By using Eq. (6) expressing the BSCs g_n^m in terms of BSCs g_n for an axisymmetric beam, we may specify E_θ and E_φ, which are then found to be

$$E_\theta = \frac{iE_0}{kr}e^{-ikr}\frac{e^{i\varphi}+Ke^{-i\varphi}}{2}\sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}g_n(a_n\tau_n + b_n\pi_n) \tag{16}$$

$$E_\varphi = \frac{-E_0}{kr}e^{-ikr}\frac{e^{i\varphi}-Ke^{-i\varphi}}{2}\sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}g_n(a_n\pi_n + b_n\tau_n) \tag{17}$$

where π_n and τ_n are Legendre functions, leading to

$$S_2 = \frac{1}{2}(e^{i\varphi}+Ke^{-i\varphi})\sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}g_n(a_n\tau_n + b_n\pi_n) \tag{18}$$

$$S_1 = \frac{1}{2}(e^{i\varphi}-Ke^{-i\varphi})\sum_{n=1}^{\infty}\frac{2n+1}{n(n+1)}g_n(a_n\pi_n + b_n\tau_n) \tag{19}$$

and to

$$|S_2|^2 = \frac{K^2+1+2K\cos 2\varphi}{8(K^2+1)}\sum_{n=1}^{\infty}\sum_{m=1}^{\infty}\frac{2n+1}{n(n+1)}\frac{2m+1}{m(m+1)}I_{nm} \times (a_n\tau_n + b_n\pi_n)(a_m^*\tau_m + b_m^*\pi_m) \tag{20}$$

$$|S_1|^2 = \frac{K^2+1-2K\cos 2\varphi}{8(K^2+1)}\sum_{n=1}^{\infty}\sum_{m=1}^{\infty}\frac{2n+1}{n(n+1)}\frac{2m+1}{m(m+1)}I_{nm} \times (a_n\pi_n + b_n\tau_n)(a_m^*\pi_m + b_m^*\tau_m) \tag{21}$$

allowing one to express the scattered intensities in terms of the polarization parameter and of the density matrix.

Next, we consider the so-called phase angle δ, which is given by [4]

$$\tan\delta = \frac{Re(S_1)Re(S_2) + Im(S_1)Im(S_2)}{Im(S_1)Re(S_2) - Re(S_1)Im(S_2)} \tag{22}$$

which may be rewritten as

$$\tan\delta = \frac{Re(S_1 S_2^*)}{Im(S_1 S_2^*)}. \tag{23}$$

Combining Eqs. (18) and (19), we obtain the quantity $S_1 S_2^*$ appearing in Eq. (23):

$$S_1 S_2^* = \frac{1-K^2+2iK\sin 2\varphi}{8(K^2+1)}\sum_{n=1}^{\infty}\sum_{m=1}^{\infty}\frac{2n+1}{n(n+1)}\frac{2m+1}{m(m+1)}I_{nm} \times (a_n\pi_n + b_n\tau_n)(a_m^*\tau_m + b_m^*\pi_m) \tag{24}$$

therefore allowing us also to express the phase angle in terms of the polarization parameter and of the density matrix.

Finally, we consider cross-sections. The scattering cross-section is [4]

$$C_{sca} = \frac{\lambda^2}{\pi}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n}\frac{2n+1}{n(n+1)}\frac{(n+|m|)!}{(n-|m|)!}\left[|a_n|^2|g_{n,TM}^m|^2 + |b_n|^2|g_{n,TE}^m|^2\right] \tag{25}$$

which, using Eq. (6), may be converted into

$$C_{sca} = \frac{\lambda^2}{8\pi}\sum_{n=1}^{\infty}(2n+1)|I_{nn}|^2\left[|a_n|^2 + |b_n|^2\right] \tag{26}$$

depending on the diagonal elements of the density matrix.

The extinction cross-section is [4]

$$C_{ext} = \frac{\lambda^2}{\pi} Re \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \frac{2n+1}{n(n+1)} \frac{(n+|m|)!}{(n-|m|)!} \left[a_n |g_{n,TM}^m|^2 + b_n |g_{n,TE}^m|^2\right] \tag{27}$$

which becomes

$$C_{ext} = \frac{\lambda^2}{8\pi} \sum_{n=1}^{\infty} (2n+1) |I_{nn}|^2 Re(a_n + b_n) \tag{28}$$

depending also on the diagonal elements of the density matrix.
In the general case, we also have to deal with three radiation pressure cross-sections Cpr, x, Cpr, y and Cpr, z. The longitudinal radiation pressure cross-section Cpr, z is [4]

$$\begin{aligned} C_{pr,z} = \frac{\lambda^2}{\pi} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \Bigg\{ & \frac{1}{(n+1)^2} \frac{(n+1+|m|)!}{(n-|m|)!} \\ & \times Re[(a_n + a_{n+1}^* - 2a_n a_{n+1}^*) g_{n,TM}^m g_{n+1,TM}^{m*} \\ & + (b_n + b_{n+1}^* - 2b_n b_{n+1}^*) g_{n,TE}^m g_{n+1,TE}^{m*}] \\ & + m \frac{2n+1}{n^2(n+1)^2} \frac{(n+|m|)!}{(n-|m|)!} \\ & \times Re[i(2a_n b_n^* - a_n - b_n^*) g_{n,TM}^m g_{n,TE}^{m*}] \Bigg\} \end{aligned} \tag{29}$$

which, in the case of axisymmetric beams, becomes

$$\begin{aligned} C_{pr,z} = \frac{\lambda^2}{8\pi} \sum_{n=1}^{\infty} \Bigg\{ & \frac{n(n+2)}{n+1} Re[I_{n,n+1}(a_n + b_n + a_{n+1}^* \\ & + b_{n+1}^* - 2a_n a_{n+1}^* - 2b_n b_{n+1}^*)] + \frac{2n+1}{n(n+1)} \\ & \times Re[I_{nn}(a_n + b_n^* - 2a_n b_n^*)] \Bigg\}. \end{aligned} \tag{30}$$

The transverse radiation pressure cross-sections $C_{pr,x}$ and $C_{pr,y}$ involve quantities denoted U_{nm}^p, V_{nm}^p, S_{nm}^p and T_{nm}^p (Ref. [4], Eqs. (147), (148), (155) and (156)), which are combinations of products of the kind $g_n^p g_n^{p+1*}$. All these products are zero for axisymmetric beams (Eq. (6)) and it is then readily observed that

$$C_{pr,x} = C_{pr,y} = 0 \tag{31}$$

as expected on physical grounds.
We note that cross-section expressions do not depend on the polarization parameter K, as expected since they are integral quantities washing out polarization effects, but only on the density matrix I_{nm} (more specifically on selected components of the density matrix). These expressions would therefore identify with those derived for on-axis Gaussian beams such as given in Refs. [5, 6]. We recall here that on-axis Gaussian beams form a subclass within the class of axisymmetric beams. The expressions for the scattered intensities and the phase angle, however, depend also on the polarization parameter, which is equal classically to 1 for on-axis Gaussian beams and therefore did not appear in Refs. [5, 6]. Therefore, the corresponding expressions given in this section, in addition to providing a density matrix approach, also generalize results of Refs. [5, 6] from the case of on-axis Gaussian beams to the more general case of axisymmetric beams.

3 Arbitrary-Shaped Beams

In this general case, the generalized amplitude functions are given by (from Eqs. (10)–(13) or explicitly from Ref. [4])

$$S_2 = \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \frac{2n+1}{n(n+1)} [a_n g_{n,TM}^m \tau_n^{|m|} + imb_n g_{n,TE}^m \pi_n^{|m|}] e^{im\varphi} \tag{32}$$

$$S_1 = \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \frac{2n+1}{n(n+1)} [ma_n g_{n,TM}^m \pi_n^{|m|} + ib_n g_{n,TE}^m \tau_n^{|m|}] e^{im\varphi} \tag{33}$$

from which we show that $|S_2|^2$ and $|S_1|^2$ can be expressed in terms of the density-matrices M_{np}^{mq}, E_{np}^{mq} and C_{np}^{mq}, according to

$$\begin{aligned} |S_2|^2 = \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \sum_{p=1}^{\infty} \sum_{q=-p}^{+p} & L_{np} e^{i(m-q)\varphi} \\ & \times \left[a_n a_p^* \tau_n^{|m|} \tau_p^{|q|} M_{np}^{mq} + mqb_n b_p^* \pi_n^{|m|} \pi_p^{|q|} E_{np}^{mq} \right. \\ & \left. - 2iqa_n b_p^* \tau_n^{|m|} \pi_p^{|q|} C_{np}^{mq}\right] \end{aligned} \tag{34}$$

$$\begin{aligned} |S_1|^2 = \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \sum_{p=1}^{\infty} \sum_{q=-p}^{+p} & L_{np} e^{i(m-q)\varphi} \\ & \times [b_n b_p^* \tau_n^{|m|} \tau_p^{|q|} E_{np}^{mq} + mqa_n a_p^* \pi_n^{|m|} \pi_p^{|q|} M_{np}^{mq} \\ & - 2ima_n b_p^* \pi_n^{|m|} \tau_p^{|q|} C_{np}^{mq}] \end{aligned} \tag{35}$$

in which we introduced

$$L_{np} = \frac{2n+1}{n(n+1)} \frac{2p+1}{p(p+1)} \tag{36}$$

therefore allowing us to express the scattered intensities I_θ^+ and I_φ^+ (Eq. (9)) in terms of the density matrices.
For the phase angle, Eq. (23) is still valid but now it is found that

$$\begin{aligned} S_1 S_2^* = \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \sum_{p=1}^{\infty} \sum_{q=-p}^{+p} & L_{np} e^{i(m-q)\varphi} \\ & \times [ma_n a_p^* \pi_n^{|m|} \tau_p^{|q|} M_{np}^{mq} + qb_n b_p^* \tau_n^{|m|} \pi_p^{|q|} E_{np}^{mq} + ib_n a_p^* \tau_n^{|m|} \tau_p^{|q|} C_{pn}^{qm*} \\ & - imqa_n b_p^* \pi_n^{|m|} \pi_p^{|q|} C_{np}^{mq}] \end{aligned} \tag{37}$$

allowing us also to express the phase angle in terms of density matrices. In particular, we may express the numerator and denominator of Eq. (23) in more symmetric forms:

$$\begin{aligned} Re(S_1 S_2^*) = Re \Bigg\{ \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \sum_{p=1}^{\infty} \sum_{q=-p}^{+p} & L_{np} e^{i(m-q)\varphi} \\ & \times \left[ma_n a_p^* \pi_n^{|m|} \tau_p^{|q|} M_{np}^{mq} + qb_n b_p^* \tau_n^{|m|} \pi_p^{|q|} E_{np}^{mq} - ia_n b_p^* \right. \\ & \left. \times \left(\tau_n^{|m|} \tau_p^{|q|} + mq \pi_n^{|m|} \pi_p^{|q|}\right) C_{np}^{mq}\right] \Bigg\} \end{aligned} \tag{38}$$

$$\begin{aligned} Im(S_1 S_2^*) = Im \Bigg\{ \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \sum_{p=1}^{\infty} \sum_{q=-p}^{+p} & L_{np} e^{i(m-q)\varphi} \\ & \times \left[ma_n a_p^* \pi_n^{|m|} \tau_p^{|q|} M_{np}^{mq} + qb_n b_p^* \tau_n^{|m|} \pi_p^{|q|} E_{np}^{mq} + ia_n b_p^* \right. \\ & \left. \times \left(\tau_n^{|m|} \tau_p^{|q|} - mq \pi_n^{|m|} \pi_p^{|q|}\right) C_{np}^{mq}\right] \Bigg\}. \end{aligned} \tag{39}$$

Next, Eq. (25) for the scattering cross-section becomes

$$C_{sca} = \frac{\lambda^2}{\pi} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \frac{2n+1}{n(n+1)} \frac{(n+|m|)!}{(n-|m|)!} \left[|a_n|^2 M_{nn}^{mm} + |b_n|^2 E_{nn}^{mm} \right] \tag{40}$$

involving generalized diagonal elements M_{nn}^{mm}, E_{nn}^{mm} of the TM density matrix M_{np}^{mq} and of the TE density matrix E_{np}^{mq}, but without any component of the cross-density matrix C_{np}^{mq}.
Similarly, Eq. (27) for the extinction cross-section becomes

$$C_{ext} = \frac{\lambda^2}{\pi} Re \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \frac{2n+1}{n(n+1)} \frac{(n+|m|)!}{(n-|m|)!} \left[a_n M_{nn}^{mm} + b_n E_{nn}^{mm} \right]. \tag{41}$$

For the longitudinal radiation pressure cross-section $C_{pr,z}$ (Eq. (29)), we obtain

$$\begin{aligned} C_{pr,z} = \frac{\lambda^2}{\pi} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} & \left\{ \frac{1}{(n+1)^2} \frac{(n+1+|m|)!}{(n-|m|)!} Re \right. \\ & \times \left[\left(a_n + a_{n+1}^* - 2a_n a_{n+1}^*\right) M_{nn+1}^{mm} \right. \\ & \left. + \left(b_n + b_{n+1}^* - 2b_n b_{n+1}^*\right) E_{nn+1}^{mm} \right] \\ & \left. + m \frac{2n+1}{n^2(n+1)^2} \frac{(n+|m|)!}{(n-|m|)!} Re[i(2a_n b_n^* - a_n - b_n^*) C_{nn}^{mm}] \right\}. \end{aligned} \tag{42}$$

In the general case, the transverse radiation pressure cross-section $C_{pr,x}$ is (Ref. [4], Eq. (159))

$$\begin{aligned} C_{pr,x} = \frac{\lambda^2}{2\pi} \sum_{p=1}^{\infty} \sum_{n=p}^{\infty} \sum_{m=p-1 \neq 0}^{\infty} & \frac{(n+p)!}{(n-p)!} [Re(S_{mn}^{p-1} + S_{nm}^{-p} - 2U_{mn}^{p-1} \\ & - 2U_{nm}^{-p}) \left(\frac{1}{m^2} \delta_{mn+1} - \frac{1}{n^2} \delta_{nm+1} \right) + \frac{2n+1}{n^2(n+1)^2} \delta_{nm} \\ & \times Re\left(T_{mn}^{p-1} - T_{nm}^{-p} - 2V_{mn}^{p-1} + 2V_{nm}^{-p}\right)] \end{aligned} \tag{43}$$

in which the quantities S_{nm}^p, T_{nm}^p, U_{nm}^p and V_{nm}^p (Ref. [4], Eqs. (147), (148), (155) and (156)) may be rewritten in terms of density matrices as

$$S_{nm}^p = (a_n + a_m^*) M_{nm}^{pp+1} + (b_n + b_m^*) E_{nm}^{pp+1} \tag{44}$$

$$T_{nm}^p = -i(a_n + b_m^*) C_{nm}^{pp+1} + i(b_n + a_m^*) C_{mn}^{p+1p*} \tag{45}$$

$$U_{nm}^p = a_n a_m^* M_{nm}^{pp+1} + b_n b_m^* E_{nm}^{pp+1} \tag{46}$$

$$V_{nm}^p = -i a_n b_m^* C_{nm}^{pp+1} + i b_n a_m^* C_{mn}^{p+1p*}. \tag{47}$$

The expression for the other transverse radiation pressure cross-section $C_{pr,y}$ is the same as Eq. (43), but with (Re) replaced by (Im), so that it can also be expressed in terms of density matrices, therefore completing the density matrix approach to GLMT.

4 Conclusion

In Part 1 [3], we demonstrated that intensity measurements on an actual beam in the laboratory should allow one to measure density matrices related to beam shape coefficients encoding the beam (and to a polarization parameter in the case of axisymmetric beams). This paper demonstrated that these density matrices are relevant for evaluating many scattering properties (scattered intensities, phase angle, cross-sections) in the framework of generalized Lorenz-Mie theory, leading to the so-called density matrix approach to the generalized Lorenz-Mie theory. The density matrix terminology has been chosen in analogy with the existence of a similar density matrix in quantum mechanics (from a formal point of view). Both the possibility of measuring density matrices and of exploiting them in generalized Lorenz-Mie theory open the way to many numerical studies and investigations on actual experiments, which are postponed to future work. Such investigations should open up new approaches in optical characterization. An actual laser beam in the laboratory may depart considerably from any ideal *a priori* theoretical description. Since optical characterization depends on scattering phenomena, and therefore on the actual structure of the illuminating beam, the interpretation of scattering experiments may be erroneous if the structure of the actual beam used is not properly implemented in the GLMT. The possibility of measuring the actual structure of the beam therefore opens the way to refined particle diagnosis.

5 Appendix – Density-Matrix Terminology

Let us consider a quantum object described by a (Schrödingerian) state ψ. The probability dP, after a measurement, of finding the particle located in a volume dV around location x is given by $\psi(x)\psi^*(x)$. More generally [7], let us consider an operator $(\hat{L})$ associated with an observable L and, referring to the theory of representations in quantum mechanics, let us consider a representation in such a way that the operator $(\hat{L})$ is represented by a matrix $L_{x'x}$. Let us also consider a pure state $\psi_\alpha(x)$. The mathematical expectation for the observable L and the pure state $\psi_\alpha(x)$ is given by

$$\bar{L}_\alpha = \int\int L_{x'x} \psi_\alpha^*(x') \psi_\alpha(x) dx dx'. \tag{48}$$

Let us then consider a set of states $\psi_\alpha(x)$ and form a mixed state in which each pure state is represented with a probability P_α. The mathematical expectation in the mixed state is then given by

$$\bar{L} = \sum_\alpha P_\alpha \bar{L}_\alpha = \sum_\alpha P_\alpha \int\int L_{x'x} \psi_\alpha^*(x') \psi_\alpha(x) dx dx' \tag{49}$$

with the proviso that we have taken $\Sigma P_\alpha = 1$. Eq. (49) may be rewritten as

$$\bar{L} = \int\int L_{x'x} \rho_{xx'} dx dx' \tag{50}$$

in which

$$\rho_{xx'} = \sum_\alpha P_\alpha \psi_\alpha(x) \psi_\alpha^*(x'). \tag{51}$$

The matrix $\rho_{xx'}$ is called the density matrix. If now the mixed state becomes a pure state $\psi_\alpha(x) = \psi(x)$ with the probability $P_\alpha = 1$, the density matrix reduces to

$$\rho_{xx'} = \psi(x)\psi^*(x) \tag{52}$$

which is to be compared with the expression for the density of the probability of presence at x.

Also, similarly, if we consider that electromagnetic fields themselves are not observable (which here means in practice that their measurement is a difficult task) and if incident beam intensities are only considered as observables, then most of the scattering quantities (scattered intensities, phase angles, cross-sections) are expressed by density matrices. More specifically, in the case of axisymmetric beams, we may consider the set of effective BSC G_ns as defining the state of the beam. Observables are then all defined by acting on the density matrix I_{nm}:

$$I_{nm} = G_n G_m^* \equiv G(n)G^*(m) \tag{53}$$

which is to be compared with Eq. (52). In the most general case where the state is defined by BSC $g_{n,TM}^m$s and $g_{n,TE}^m$s, we need three different density matrices, without, however, spoiling the interest of the (formal) analogy.

6 Acknowledgements

During the study of the possibility of measuring BSCs or associated density matrices, a continuous fruitful and enjoyable interaction occurred with Professor *J. A. Lock* of Cleveland University. It was he who suggested the density matrix terminology. I found it splendid and adopted it straight away.

7 Symbols and Abbreviations

a_n, b_n	scattering coefficients
BSC	beam shape coefficient
C_{np}^{mq}	cross-density matrix
C_{sca}	scattering cross-section
C_{ext}	extinction cross-section
$C_{pr,x}, C_{pr,y}$	transverse radiation pressure cross-sections
$C_{pr,z}$	longitudinal radiation pressure cross-section
E_i	electric field components
E_0	electric strength
E_{np}^{mq}	TE density matrix
g_n	beam shape coefficients for axisymmetric beams
G_n	effective beam shape coefficients (axisymmetric beams)
GLMT	generalized Lorenz-Mie theory
$g_{n,TM}^m, g_{n,TE}^m$	beam shape coefficients for arbitrary beams
I_{nm}	density matrix (axisymmetric beams)
I_θ^+, I_φ^+	scattered intensities
k	wavenumber
K	polarization parameter
$\hat{L}$	operator in quantum mechanics
L	observable associated with operator $\hat{L}$ in quantum mechanics
$\bar{L}$	mathematical expectation in a mixed state
L_{np}	Eq. (36)
$L_{x'x}$	matrix representation of the operator $\hat{L}$
$\bar{L}_\alpha$	mathematical expectation for the observable L and a pure state ψ_α
M_{np}^{mq}	TM density matrix
P_n^m	associated Legendre polynomials
P_α	probability of a pure state ψ_α in a mixed state
S_1, S_2	generalized amplitude functions
(r, θ, φ)	spherical coordinates
(x, y, z)	Cartesian coordinates
δ	phase angle
δ_{nm}	Kronecker symbol
ϵ_0	electric capacity of free space
π_n, τ_n	Legendre functions
π_n^m, τ_n^m	generalized Legendre functions
$\rho_{x'x}$	density matrix in quantum mechanics
μ_0	magnetic capacity of free space
ω	angular frequency
ψ	state function of a particle in quantum mechanics
ψ_α	pure state function
λ	wavelength

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