

Generalized Lorenz-Mie Theories

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Et Dieu dit: “Voici le signe de l’alliance que je mets entre moi et vous, et tous les êtres vivants qui sont avec vous, pour toutes les générations à venir. Je mets mon arc dans la nue, ce sera un signe d’alliance entre moi et la terre. Quand j’assemblerai des nuées au-dessus de la terre, l’arc apparaîtra dans la nue.”

Gn, 9, 12-14. La Sainte Bible du
Chanoine Crampon

Schwüles Gedünst schwebt in der Luft;
lästig ist mir der trübe Druck:
das bleiche Gewölk
samml'ich zu blitzendem Wetter;
.....
Weise der Brücke den Weg!

Richard Wagner.
Das Rheingold
Vierte Szene.

Preface

Examining the general subject of light scattering is like examining a gemstone having many facets. As a result, when people talk about light scattering they can mean any one of many things. Light can be scattered by individual particles of high symmetry (Lorenz-Mie theory or LMT) ranging in size from a small fraction of the wavelength of light (Rayleigh scattering) to many thousands of wavelengths (semiclassical scattering and ray theory), or by irregularly-shaped particles (e.g. Null-Field method). People can also mean scattering of light by crystalline or amorphous aggregates of such particles (Bragg scattering or pair-correlated scattering), or the repetitive scattering of light through a dense cloud of particles (multiple scattering and radiative transfer). People can mean scattering of light from sound waves in a gas (Rayleigh-Brillouin scattering), from thermally generated capillary waves on the surface of a liquid (surface scattering), or from long range density fluctuations in a system undergoing a phase transition (critical opalescence). They can mean the Doppler shift of the frequency of the light scattered by particles entrained in a flow (quasi-elastic light scattering), or the time dependent interference of light scattered by many particles undergoing Brownian motion (dynamic light scattering and diffusing wave spectroscopy). They can also mean the absorption of light by the molecules of a particle and re-radiation at a lower frequency (inelastic scattering). Since light scattering is such a large area of endeavor, one needs to clearly state which of the many facets one will be describing when explaining it to someone else. The particular facet that is the topic of this book is scattering by individual particles having a high degree of symmetry (LMT) by a transversely focused beam, which is today called generalized Lorenz-Mie theory, or GLMT for short.

There once was a time when the world of Lorenz-Mie scattering was relatively simple. LMT had been developed by a number of researchers during the time period extending from the last few decades of the 19th century to 1908. It provided an exact solution of the electromagnetic boundary value problem of scattering of an incident plane wave, such as sunlight, by a dielectric spherical particle, such as a small water droplet. The solution took the

form of an infinite series of partial wave terms. This was the worst possible type of exact solution to have since the individual terms were cumbersome to calculate and the infinite series was frequently extremely slowly convergent. Unfortunately, in spite of these limitations, it was the only form of the exact solution to the light scattering problem that was known. Some researchers contented themselves by studying scattering from particles that were small compared with the wavelength of light, where the series contained only one term. Others devised clever analytical approximations such as Airy theory, asymptotic expansions, and stationary phase methods to describe scattering by a raindrop which was hundreds or thousands of times larger than the wavelength of light, and where the infinite series converged only after many hundreds or many thousands of terms. Yet others used motor-driven mechanical calculators until they overheated to calculate the ten or twenty terms in the series that were required for convergence when the water droplet was only a few times larger than the wavelength of light. By the early 1960's the computational difficulties had improved in the sense that those who had sufficient grant money to purchase run time on a mainframe computer could calculate Lorenz-Mie scattering for any size particle, and produce either long tables or large collections of graphs of the scattered intensity as a function of angle for various types of particles.

The invention of the gas laser in the early 1960's and the popularization of personal computers in the 1970's changed everything. Now anyone with a laser, a few lenses, a photodetector, and a PC could study light scattering and use it as a particle characterization tool. But, the nature of the laser beam itself caused everyone to realize that the trusted and time-tested Lorenz-Mie theory could no longer adequately describe what was now being observed in the laboratory. The width of the laser beam could easily be focused down to the diameter of the test particle or less, whereas Lorenz-Mie theory had assumed that both the beam amplitude and phase were constant over the particle diameter. In addition, people were very excited that all the things one could only dream of calculating back in the old days of mechanical calculators and mainframe computers could now be easily calculated if only one had the appropriate theory. If one had a PC, then run time was now free no matter how many hours or days the calculation took using the 2MHz clock speed and 64K of memory available in the first generation of machines.

And so the race was on to find a new extension of Lorenz-Mie theory appropriate to lasers. What theory of electromagnetic scattering by a tightly focused laser beam could be devised that was exact but also would be practical to use? After a number of initial theoretical attempts that ranged from somewhat unrealistic to somewhat successful, it began to become apparent that what today is known as GLMT might well be the best bet for a theory that was both mathematically exact and practical to implement. Practical is now defined differently than in the past due to both continuing improvements in memory and speed (but not price) of PCs, and their increased availability.

Beginning in the late 1970's, the epicenter of the development of GLMT was located at the University of Rouen in the light scattering group of Prof. Gérard Gouesbet who headed the theory section and Dr. Gérard Gréhan who headed the experimental section. The continuous interplay between theory and experiment at Rouen kept the theory honest, maintaining the theoretician's dream of developing a theory that was mathematically exact while conforming to the experimenter's demand that the theory be practical to use. This book, written by the two Gérards who were centrally involved in the development of GLMT from the very beginning, tells the story of the theory both traveling down the main road of its development as well as down a number of side roads that discuss specific technical details. The development of GLMT was the product of many researchers both in Rouen and elsewhere working over a long period of time. A reasonably complete list of these participants is given immediately before the Introduction, and their contributions are amply referenced in Chapters 3 through 7. As one of these participants, I can say from personal experience that all the results that seem so obvious now, and the progressions of ideas that seem so straightforward, were not so obvious or straightforward then. Much soul searching, serious debate, and worry about how bold to be in print were required to develop GLMT from where it was in those years to where it is today.

The basic idea of GLMT is that a transversely localized beam that is a solution to Maxwell's equations can be written as an infinite series of spherical Bessel functions and spherical harmonics, each multiplied by a coefficient that is called a beam shape coefficient. There are then two separate parts to the derivation and application of the formulas of GLMT, (i) the way in which all the formulas of experimental interest depend on the beam shape coefficients, and (ii) how the beam shape coefficients are calculated for a particular beam of interest. The first part was understood from almost the very beginning. But, the second part turned out to be where all the complications were lurking, and it took many years to sort these complications out. The basic results from electromagnetic theory necessary to derive the main formulas of both LMT and GLMT are recounted in Chapters 1 and 2, and the GLMT formulas of experimental interest are written in terms of the beam shape coefficients in Chapter 3. This gives the overall essence of GLMT, except for the one nagging question that turned out to be a Pandora's box. Namely, how does one determine the beam shape coefficients for what people call a Gaussian laser beam, or some other type of beam? This question required a study, in much more detail and to a much greater depth than anyone could have realized beforehand, of exactly what a focused beam is and what it means for a beam to be an exact solution of Maxwell's equations. This was a great struggle because people knew that before one can decompose a beam into an infinite series of spherical Bessel functions and spherical harmonics, one has to know exactly what the beam is that one is attempting to decompose. It turned out that what people colloquially called a Gaussian beam was not in actuality a solution of Maxwell's equations. By the act of determining the

beam shape coefficients of such a beam, one remodeled it from its original shape that was not quite a solution of Maxwell's equations to another beam that had not quite the original shape but that was a solution of the equations. It took a long time to understand this and to suitably control the beam remodeling procedure, even though in retrospect it is now all clear enough. This is why the various methods for obtaining the beam shape coefficients are spread out over Chapters 4-7. After all this effort over many years, I would not be surprised if the last word on obtaining and understanding the beam shape coefficients has yet to be written. But, the current state of the art is good. It is certainly able to be implemented in a practical and reliable way, and it agrees nicely with the results of experiments.

Perhaps a measure of GLMT's maturity today is the fact that although there are still new theoretical developments related to fundamental physics and mathematics, the subject is no longer dominated by them. GLMT has instead become a useful and valued tool for engineers who wish to characterize small solid or liquid particles and use these scattering measurements to assist in their design, testing, and calibration of a large variety of products, devices, and instruments. Many applications of GLMT are described in detail in Chapter 8 and are generously referenced there. As a similar measure of the maturity of the theory, although heroic individual efforts are still being made in code generation for various exotic extensions and uses of GLMT, both Mie and GLMT codes have become robust and highly developed. Standard libraries of them do exist, they have been published in books, and they are now available on the Internet. A highly developed and greatly tested library of computer programs is provided in the website connected to book.

I believe this book on GLMT has been written by the two Gérards to serve a large variety of audiences. On the one hand, with its computer programs it is useful as a practical tool for those who wish to apply GLMT to the interpretation of laboratory measurements. On the other hand, since the derivations in the book are rather complete with only a few steps left out here and there, the book serves as a useful archival reference for students seeking to learn the mathematics and physics of the subject. On yet a third hand, since most of the derivations begin in rather complete generality and many specific theoretical fine-points are discussed in detail, the book provides a good starting point for advanced researchers interested in either developing new insights into GLMT or extending it to particles having a more complicated response to external electric and magnetic fields. Since GLMT was developed largely by the Rouen group headed by Prof. Gérard Gouesbet and Dr. Gérard Gréhan, and since the two Gérards had leading roles in both the theoretical development and experimental testing of the theory from the very beginning, it is only natural that this book is authored by them.

Many hundreds of years ago it was believed that knowledge had reached its peak under the ancient Greeks. When one was studying a certain natural phenomena in those times, one would always ask what Aristotle, the greatest authority on all of Natural Philosophy, had to say on the subject. As I was

preparing to write this preface, I read the prefaces of a number of other specialized books on various facets of light scattering to see what other people had written. In doing so I came upon a quote I had never seen before. In the book “Light Scattering by Nonspherical Particles” edited by Michael Mishchenko, Joachim Hovenier, and Larry Travis (Academic Press, 2000) there is a Forward written by H. C. van de Hulst (pp.xxv-xxx). Near the end of the Forward, van de Hulst writes

“In more recent work on Mie theory two developments please me most:

(a) The many papers, mostly by Gouesbet and his co-workers, on scattering of a focused (laser) beam that illuminates the sphere eccentrically

(b) The glare points (in some papers wrongly called rainbows) showing under which angles the most intense radiation exits from a sphere fully illuminated by a distant source”

While I do not intend to place Henk van de Hulst on the same pedestal as Aristotle, this is a very nice tribute to GLMT nonetheless. In a similar vein, one might also parenthetically conjecture what the G might possibly otherwise be an abbreviation of in the acronym GLMT.

January 2010

James A. Lock
Cleveland, Ohio, USA

Preliminaries

This book is written in English, or may be in American, or more likely in some kind of international language mixing miscellaneous influences from various countries. Whatever the used language is, it is not the natural language of the authors. Furthermore, because this book is written (that is to say not spoken), the reader will miss the delicious Frenchy Maurice Chevalier accent which is a part of our charm: nothing is perfect in this poor world!

Actually, most of our writing has been checked by our friend and colleague Alain Souillard, from the Rouen National Institute of Applied Sciences. However, such a checking is not enough to ensure correctness because:

- (i) several versions of each chapter have been written before approaching the final asymptotic version and it would not have been friendly to ask Alain Souillard to check again and again
- (ii) scientific terminology has not been checked just because Alain Souillard is not a scientist

At least, we believe that the reader will forgive us for the use of gallicisms and remaining language incorrectnesses. They should not be strong enough to prevent sufficient understanding.

We feel more concerned by the fact that misprints and incorrectnesses in formulae are likely to occur in such a book. Indeed, even a sign error may be a nightmare to theoreticians. As a relief, we may state that such problems are a manifestation of a physical reality, namely the creation of entropy along an information channel. Although this is however a very small relief indeed, the only remaining way to escape from the shame of making errors is to state that the blame for remaining imperfections rests on the shoulders of the other author, or on Nature which made us.

We are also looking forward to the readers who will kindly help us to improve the book by communicating to us regarding any incorrectness. Thanks to them in anticipation.

Acknowledgments

The development and completion of the generalized Lorenz-Mie theory (GLMT) *stricto sensu* and, more generally, of the generalized Lorenz-Mie theories (in the plural, as we shall explain later), and of applications, took place at the University of Rouen and at the National Institute of Applied Sciences of Rouen, first on the campus of Mont-Saint-Aignan and, thereafter, on the campus of Saint-Etienne du Rouvray. Very likely (as far as we remember), the first equations have been written in 1978 and the first article paper has been published in 1982 [1]. Therefore, more than thirty years have elapsed since the beginning of the story, and the third decade is over.

During this period of time, many students contributed to the topic, and it is here a right place and a right time to acknowledge all of them, namely (using a chronological order) : B. Maheu (who helped taking care of the first chapter of this book), F. Corbin, F. Guilloteau, K.F. Ren, F. Onofri, D. Blondel, H. Mignon, T. Girasole, C. Rozé, N. Gauchet, H. Bultynck, H. Polaert, S. Meunier-Guttin-Cluzel, L. Mées and J. Ducastel. During such a long time, we also had many opportunities to enjoy local collaborations (using an alphabetic order) : D. Allano, S. Belaid, M. Brunel, D. Lebrun, E. Lenglard, D. Lisiecki, C. Ozkül, F. Slimani, national collaborations : M.I. Angelova, D. Boulaud, P. Cetier, J.P. Chevaillier, J.B. Dementon, J. Fabre, K.I. Ichige, A. Kleitz, G. Martinot-Lagarde, B. Pouligny, F. Vannobel, J.P. Wolf, and international collaborations : G. Brenn, X. Cai, N. Damaschke, J. Domnick, F. Durst, L.X. Guo, Y.P. Han, P. Haugen, J.T. Hodges, R. Kleine, J.Y. Liu, J.A. Lock, J. Mroczka, A. Naqwi, Y. Ohtake, C. Presser, V. Renz, B. Rück, H.G. Semerjian, X. Shen, I. Shimizu, Y. Takahara, A.K.M.P. Taylor, C. Tropea, A. Ungut, Y. Wang, I. Wilhelm, Z.S. Wu, D. Wysoczanski, F. Xu, T.H. Xu, S. Zhang, M. Ziema, and to make friends. We also acknowledge R. Petit from Aix-Marseille University who kindly accepted to check the first chapter and helped us to improve it.

From the first equations to what we consider as the “pivot” article in 1988 [2], ten years were required. Another five years has been necessary to reach the first genuine applications of the theory to a concrete problem in optical particle characterization, namely concerning the issue of trajectory ambiguity effects in phase Doppler anemometry [3], [4]. This was more than sufficient to pile up several thousands of pages of computations, including the ones spent in blind alleys, and the time spoiled by jumping cliffs or producing rubbish stuff. All this material has been preserved as a testimony of madness. As one can guess, some people wondered whether we were actually serious or not, crazy or not, what this theory was made for, whether it would be useful or not, worthwhile or not, or whether it was simply a game boosted by mind perversion. We therefore experienced some kind of negative social pressure which actually contributed to the expansion of the universe of GLMT. Anonymous acknowledgments are also required in this matter.

Introduction

For a physicist, and more generally for a scientist, one of the pleasures in everyday life is to detect exemplifying manifestations and avatars of the topic he is working on. Why do small tea leaves cluster to the center of the free surface of liquid in the cup, contrasting with the fact that centrifugal forces should drive them towards the wall? Why is liquid poured outside of a pot usually keen to follow the pot wall onto the table, in a persistent and irritating way, rather than flowing down, more or less vertically, to the cup? Why does a mirror change left to right but not top to bottom?

These three examples clearly concern tiny facts but should not be looked down upon when thinking about more challenging questions of physics and metaphysics, for small lanes may lead to large and unknown avenues. Actually a fascinating aspect of science is that it permits the study of familiar phenomena which are universal to some extent in so far as every human being may experience them, even if one stays away from modern cathedrals, we mean laboratories. Furthermore, behind these familiar phenomena are hidden deep arcana to decipher and reveal. Behind the simple action of shaking an ink-pen to supply ink to the pen, there is inertia and classical mechanics, while quantum theory is written on the sky by lasers piercing the night in some contemporary shows.

In optics, and to stick more closely to the topic of this book, in light scattering by particles, everyday surprises and observations may be specially appealing because they concern what could be the most privileged sense of humankind, namely vision. This metamorphoses photons to an enchanting world of shapes and colours that nearly everyone may enjoy, very often with a feeling of infinity and eternity, for instance when the sun rises on the Mississippi river or falls gently to sleep on Fuji-San.

The most striking example might be the rainbow, so marvelous that it has become a signature at the bottom of a contract between God and humanity in Jewish and Christian religions, or the scarf of Goddess Iris delivering messages from heaven to earth in the Greek mythology, or a bridge to Walhalla in 'Der Ring des Nibelungen' by Richard Wagner, inspired by old germanic and

nordic legends. Even questions as naïve as : "Why is the sky blue at day while it displays all shades of reds in some dawns and twilights?", and "Why is it not as bright as the sun at night?", may lead to significant research and conclusions.

To give some qualitative clues for answering the first question, we mention that aforementioned blue and reds are due to the scattering of solar light by molecules and particles of terrestrial atmosphere. Moreover, scattering plays an essential role in cloudy days (a rather common feature in our Normandy), and also in foggy days (not so common, but still far from being exceptional). During these days it is possible to walk and drive without any infra-red assisting device thanks to a significant amount of solar and/or headlights photons which are scattered by droplets and crystals and somehow succeed to reach landscape details where they are scattered again to eventually reach the eyes. More generally, scattering is essential to our vision of the world since things are only visible thanks to the light they scatter from natural or artificial sources, changing the direction of travelling photons, modifying their amount, and generating colours through complex phenomena. As far as the above second question is concerned, it is deeply connected with the cosmological issue of the finiteness of Universe in space-time besides being may be a subtle coincidence allowing our eyes to get some rest!

In astronomy, the study of the light scattered by planetary atmospheres provides information on their composition ([5], [6]) but zodiacal light scattered outside of the ecliptic plane by dust particles of the solar planetary system limits the performances of spatial telescopes. In medicine, scattering of light by red cells of the blood enables to determine oxygen concentration ([7]), and the detection of tooth decay can be accomplished by studying the light scattered by tooth enamel ([8]). In surgery, there is interest in studying scattering interactions between light and biological tissues in order to master laser surgery as well as possible ([9]). In industry, applications of light scattering are manifold and potentially infinite in number. We would need a scientific and reckless Prévert to attempt to catalogue an endless inventory. Being scientists, but neither the reckless nor Prévert's type, we shall be content to mention some examples including the control of the transparency of drinking glasses, and of the luminous and mechanical properties of paints and sheets which depend strongly on the embedded scattering particles ([10], [11], [12], [13], old classical articles indeed). Other specific examples more relevant to the purpose of this book will be given later in a more restricted context.

This more restricted context is the electromagnetic scattering by particles.

Concerning scattering, our framework is Quasi-Elastic Scattering (QES) which refers to the case when no change of frequency is involved in the light/matter interaction except that one due to the Doppler effect and the other singular one, from a finite frequency to a null frequency, when a photon is absorbed. Also, although it may be convenient to think of light in terms of photons and although scattering is a random quantum process at a more

fundamental level of description, assemblies of photons will be here modelled as electromagnetic waves.

Concerning particles, they *a priori* might be of arbitrary shape, arbitrary nature, arbitrary size, and embedded in media of arbitrary geometry with arbitrary distribution of particle concentrations.

The induced complexity is however still too big for us to handle in this book. The reader is invited to go to famous studies which should be present on any library shelf of a decent "scatterist" to get extensive information on the QES and underlying electromagnetism, such as those quoted as [14], [15], [16], [17], [18], [19], [20], [21] and [22]. As far as we are here concerned, attention will be focused on more restricted topics, not in an arbitrary way but because of the overwhelming difficulties involved in attempting to handle arbitrary situations with a single non arbitrary theory.

A first dichotomy to introduce might be between multiple and single scatterings. If it is temporarily accepted to describe light in terms of photons (whatever they are), then single scattering takes place when a photon entering a medium leaves it without having suffered more than one proper scattering event or when the first interaction between the photon and one particle leads to absorption. If more than one event (for instance two successive scatterings or one scattering followed by absorption) are involved during the life of the photon in the medium, then we are faced with multiple scattering. When the particle concentration is high in the medium, more complex phenomena may occur like dependent or coherent scattering (for instance, [23]). For multiple scattering, the interested reader may refer to the beautiful books by S. Chandrasekhar [24] and H.C. van de Hulst [18], and also by G. Kortüm [25] who introduces simple and efficient ideas in an appealing way. However, only single scattering will be here discussed (for most of the time) and it will be difficult enough to be enjoyable.

A second dichotomy is between direct and inverse problems. Assume that a particle is illuminated by a plane wave or by a laser beam and that you need know the properties of the scattered light. This is a direct problem. Conversely, assume that you know the properties of the scattered light but that you need the properties of the particle. This is an inverse problem. This dichotomy holds actually for both multiple and single scatterings. Rather clearly, an inverse problem is more difficult to solve than a direct problem. This increase of difficulty is vividly illustrated by Bohren and Huffman [22](pp. 9-11) who stated that a direct problem consists in describing the tracks of a given dragon while the inverse problem is to describe the dragon from its tracks. Most of this book will be devoted to direct problems, but optical particle sizing and, more generally, optical particle characterization, discussed in the last chapter, point out to genuine inverse problems.

Now, let us zoom again on a more restricted part of the landscape. Media containing a huge number of particles are essentially outside of the scope of this book. Suffice it to say that when particles are randomly distributed in space, it is usually enough to forget any phase relations and interference

phenomena and simply to sum up scattered intensities to solve the direct problem. Therefore, we are here mostly concerned with the interaction between an electromagnetic wave and a single particle (or scatterers made out from a small number of particles, with coherent scattering).

As far as the particle is concerned, it can be of arbitrary size (not too small however to still own a bulk macroscopic character, and reasonably not too large since particles of infinite dimension do not exist) but its nature and shape are carefully chosen to make life easy, i.e. they are regular particles allowing one to use a method of separation of variables.

Most of the book is however devoted to the case when the scatterer is a homogeneous sphere defined by its diameter d and by its complex refractive index m . Then, when the incident wave is an usual ideal plane wave, the problem was actually solved about one century ago. The corresponding theory is usually granted to G. Mie [26]. However there is certainly some injustice in that. We must remember the work that Lorenz (often mis-written as Lorentz) accomplished about twenty years before ([27], [28]) although, admittedly, Lorenz is not likely to worry any more about it (but who knows actually?). The reader could refer to an article by N.A. Logan [29] to understand how Lorenz work has been unfairly overlooked, and also to historical articles from the 2nd International Congress on Optical Particle Sizing ([30] [31] and [32]). Even if an important Lorenz memoir has been lost, it still remains that he solved the problem of the scattering of waves by dielectric spheres though without explicitly referring to Maxwell's equations. Hence, it could be recommended to speak of the Lorenz-Mie Theory (LMT) instead of simply the Mie theory. One year after Mie, Debye ([33]) completed the theory by discussing the radiation pressure. Some people might then recommend to speak of Lorenz-Mie-Debye theory, but we feel that it is unnecessary. Debye being also very well known for other contributions to physics, he has already got his full share of fame.

Now, there is more to tell concerning the relationship between Mie and Lorenz. While Mie indeed relied on the macroscopic version of Maxwell's electromagnetism, Lorenz conversely relied on a mechanical theory of aether. Yet, both theories are empirically equivalent. The inquisitive reader might amazingly wonder how a "correct" theory (presumably the one based on Maxwell's equations) could agree with an "erroneous" theory (presumably the one based on mechanics). This is a very deep epistemological issue that cannot be extensively developed in this book. It is certainly an example of what is sometimes called the Duhem-Quine theorem telling that theories are under-determined by experiments [34], [35], [36], [37], [38].

One of the authors (Gouesbet Gérard) would now like to come to the original motivation which led to the theoretical developments described in this book. During his state thesis (a kind of thesis pertaining to a previous French university system), he studied diffusion and thermal diffusion phenomena of neutral species in a plasma of argon and helium [39]. This study required the measurements of plasma velocity by using laser Doppler velocimetry which,

nowadays, is a well established technique for measuring flow velocities. In the most current optical set-up, a set of (more or less) parallel fringes is generated by making two laser beams (originating from the same source) interfering in a control volume. The flow under study is supposed to transport small inertia particles called tracers. When such a particle crosses the control volume, it generates a scattered light which is modulated at a certain frequency depending on the fringe spacing and on the velocity of the tracer, that is to say on the velocity of the flow (more properly, on the velocity component perpendicularly to the fringes). A photodetector is used to produce an electronic signal which is afterward processed by a processing device.

The plasma aforementioned above was a high frequency (laminar) plasma whose atom temperature was typically equal to 5 000 K (and the electron temperature to typically 10 000 K). It was seeded with alumina particles. The high temperatures involved led to a dilemma. If the injected alumina particles were too small, then they vaporized and could not produce any Doppler signal. If they were too big, then they would not be tracers any more, drifting behind the plasma flow. A compromise was necessary but there was no way at that time to control this compromise. The validity of the velocity measurements could only be indirectly checked (by verifying the conservation of mass), but there was no direct available way to do it. Furthermore, to make the situation a bit more confused, there was an erroneous dogma at that time (to become erroneous is common for a dogma) according to which decent Doppler signals could only be produced by particles smaller than the fringe spacing. However, it soon became obvious that having good Doppler signals did not imply that the particles were smaller than the fringe spacing, that is to say that they could be assumed to genuinely behave as tracers [40], [41]

The best direct way to solve the problem would have been to possess an instrument allowing one to measure simultaneously the size and the velocity of individual particles in flows, not only for small particles (tracers) but also for large particles. Similar needs and questions were put forward in different fields, like in plasma spraying or in the study of sprays in combustion systems. Several systems were proposed and studied, like relying on the use of the visibility or of the pedestal of Doppler signals [42], [43]. But, although satisfactory results were published in the archival literature or announced in conferences, there, however, were also many people becoming disappointed up to a situation where the topic of optical sizing received a rather poor reputation. In particular, the visibility and the pedestal techniques are no longer used nowadays. It became obvious that the topic was too much based on experiments, somewhat of a nearly pure empirical nature, without a sufficient theoretical effort to master the design and functioning of instruments.

What is likely to be the most important problem is that, in optically measuring the sizes of discrete particles in flows (possibly simultaneously with velocities, complex refractive indices, and concentrations), a laser source is usually required or, at least, very useful. Under some circumstances, the laser beam is expanded and/or the particles to be studied are small enough,

in such a way that the laser source can be safely considered as being a plane wave. An example is provided by the diffractometry technique. To design instruments and to interpret data, one can then rely more or less blindly on the basic theory for plane wave scattering, namely LMT. Indeed, for a long time, LMT has been the most powerful theoretical tool in that respect and, even nowadays, it remains famous and is being used. However, this theory is now a one-hundred-year-old lady, and although still very alive and waltz dancing, it is no wonder that it might become inappropriate in a large variety of situations, since the laser was still in the nimbus when Ludwig Lorenz and Gustav Mie write their equations.

Very often, the situation is not ideal enough to plaster an old theory on contemporary experiments. In LDA-based systems for instance, most data processing, design of instruments, and theoretical principles rely on the classical LMT, although the in going laser beams are usually focused. When the diameter of the discrete particles is not small with respect to the laser probe diameter (usually a Gaussian beam diameter or the width of the plateau in a top-hat beam), then we have to worry on the validity of the LMT which could be misleading. Also, in some cases, measurements directly rely on laser beam scattering properties ([44]). Consequently, we certainly need to rely on a more general theory, enabling us to compute the properties of the light scattered by an ideal sphere illuminated by a Gaussian beam or a top-hat beam, more generally by an arbitrary-shaped beam.

We shall call this theory the generalized Lorenz-Mie theory (GLMT), or sometimes the generalized Lorenz Mie theory in the strict sense (*stricto sensu*) to which most of this book is devoted. Nevertheless, we shall also consider other cases, when the scatterer is not a homogeneous sphere defined by its diameter d and its complex refractive index m , but is another kind of scatterer whose properties allow one to solve the scattering problem by applying a method of separation of variables. The terminology "generalized Lorenz Mie theory" will be also used for these other cases, with the proviso that the kind of scatterer under study has to be specified. For instance, there will be a generalized Lorenz-Mie theory for infinite circular cylinders.

The existence of GLMTs is relevant to the understanding and to a better design of optical measurement techniques, such as for simultaneous measurements of velocities and sizes of particles embedded in flows, and therefore to the study of multiphase flows transporting discrete particles. Two-phase and multi-phase situations of this kind (suspensions, droplet and bubble flows) are indeed very common in industry, laboratories and environment. Examples involve the control, understanding and design of specific spray and particle combustion systems, of laden flows encountered by chemical and mechanical engineers in pipes and conduits, or also in standard chemical engineering processes such as fluidization, sedimentation and pneumatic transport. People may also be concerned with very small particles such as soots which are deeply inhaled into the lungs and can lead to the development of tumours, exemplifying that environmental care is one reason to develop optical particle

sizing techniques (more generally optical particle characterization), in order to learn how to control the emission and the dispersion of such particles. Particularly relevant to this topic is the use of Diesel engines in individual cars, where we have to worry about the effects of exhaust particulate emission. In connection with energy supply and more precise control of energy conversion devices, we also have to perform characterization of droplets in spray flames and of solid particles in coal pulverized flames. Hydraulic engineers are concerned with sand and pollutant transport in rivers, lakes, and also in oceans where one must investigate spray droplets over the sea or deposition and dragging away of particulates in the sea and on the ground in connection with water motion. Industrial processes also include particulate clean-up devices such as electrostatic precipitators, oil mists from pumps, the influence of particle properties for cements and paints. These heteroclit examples are obviously very far from providing an exhaustive list, and only aim at giving a flavour of the richness of possible applications.

From a fundamental point of view, the knowledge of the size and the velocity distributions (or better of the size and velocity, particle by particle) of a dispersed phase is relevant to the understanding and to the prediction of heat and mass transfer, and of chemical reactions in many processes such as in combustion systems. It is also relevant to the understanding and to the prediction of the dispersion of particles by continuous motions in turbulent flows, a domain of research which is very active nowadays.

Therefore, the study of two-phase flows in which discrete particles are transported in and by turbulent structures are of interest both to the researcher, wishing to understand and describe the laws of nature, faced with difficult and challenging two-phase flow problems, and to the engineer who inevitably encounters them in a large variety of industrial situations, and has to design and control various plants and processes involving particle heat and mass transfer phenomena.

At first, the engineer may rely on correlations based on experimental results and adequate display of data using pertinent dimensionless groups. However, correlations are usually valid only for limited ranges of parameters, and extension of results beyond these ranges is always a risky affair. Then, the researcher must open the way for a second step, namely predictions through modelling and computer programming. This is also a risky affair which requires careful validations against well-designed experimental test-cases. For a background in such problems, see [45] [46], [47], [48] [49] [50] and references therein.

Consequently, in any case, accurate and extensive measurements of sizes, velocities, and concentrations (and may be also shape and refractive index characterization) of the discrete particles in the flow under study are needed. The ideal aim would be to provide us with space- and time-dependent particle size spectra. Emphasis must be set on optical techniques which may be non intrusive (in principle they do not disturb the medium under study), and

supply us with local (in situ small control volumes), time-resolved data. A very relevant key-word for these techniques is: versatility.

Significant advances have indeed been accomplished for years thanks to the development of these optical techniques, combining laser at the input and computer at the output, two requisites of numerous modern experiments lying somewhere within light scattering theory.

The proceedings of a series of symposia, hold under the name : "Optical particle sizing : theory and practice" later on generalized to "Optical particle characterization", would allow the interested reader to follow the development of the field in a very comprehensive way. After the first symposium in Rouen, France, in 1987 organized by the authors of the book, subsequent conferences have been hold in Phoenix, Arizona, in 1990 (chaired by D. Hirleman), in Yokohama, Japan, in 1993 (chaired by M. Maeda), in Nüremberg, Germany, in 1995 (chaired by F. Durst), in Minneapolis, Minnesota, in 1998 (chaired by A. Naqwi), in Brighton, England, in 2001 (chaired by A. Jones), in Kyoto, Japan, in 2004 (chaired by M. Itoh) and in Gräz, Austria, in 2007 (chaired by O. Glätter). Beside proceedings *stricto sensu* e.g. ([51], [52], [53], [54]), selected articles have been published in "Applied Optics" and "Particle and Particle Systems Characterization".

Actually, although we had a specific motivation in mind when developing the GLMT, as discussed above, it is now clear that the range of applications has extended far more beyond what was originally expected and, let us tell it frankly, toward unexpected fields. An example concerns the interpretation of optical levitation experiments, see e.g. [55], [56], [57], [58], and references therein. However, many other applications have to be discussed, and they will indeed be discussed.

Discussions of scattering from shaped beams have previously been provided by several authors, with more or less extended degrees of generality. Indeed, the development of science takes place through a filiation where nothing emerges from nothing. We now briefly but fairly exhaustively discuss these works, limiting here ourselves to the ones prior to 1989. We start with Chew *et al* [59] [60] who discuss converging and diverging beams (respectively). To approach the case of laser beams, Morita *et al* [61] assume a beam with a Gaussian distribution of amplitude (which does not comply with Maxwell's equations however) and a scatter center smaller than the waist and located near it. Tsai and Pogorzelski [62] consider a TEM_{00} beam which is described by using an expansion of vectorial cylindrical functions in vectorial spherical functions. Then, the beam description complies with Maxwell's equations but exhibits two singularities where the electric fields are zero, located at $\sqrt{2}$ times the waist radius from the beam axis. Their results concern small scatter centers centered at the beam waist. Tam [63] and Tam and Coriveau [64] generalize the method of Tsai and Pogorzelski to the beam mode TEM_{01}^* and to arbitrary location of the scatter center but do not present any numerical results. There is also a series of articles co-authored by Yeh ([65], [66], [67]) starting from a Rayleigh-Gans approximation up to the 1982-article in which

the TEM_{00} beam is described by its plane wave spectrum and the location of the scatter center is arbitrary. However, particles must be non absorbing and a practical limitation results from the time-consuming character of numerical computations (published results only concern particles with a size parameter smaller than about 5, i.e. diameters smaller than about twice the wavelength of the incoming electromagnetic wave). Kim and Lee [68] [69] describe the incoming TEM_{00} laser beam by using a complex point-source method and establish the corresponding theory for a spherical scatter center arbitrarily located in the beam. However, the complex point-source method introduces two singularities at which field amplitudes become infinite. Finally, we mention a more recent study by Barton *et al* [70]. Barton contribution will be given more discussion when appropriate in this book.

With the first GLMT-equations likely written in 1978, the first releases of the existence of a GLMT are testified (*urbi*) in the PhD thesis of G. Gréhan, in 1980 [71] or, the same year, in an internal report [72] and thereafter (*orbi*) in an AIAA conference in Palo Alto, California, in 1981 [73]. The first archival article, in which known precursors were acknowledged, was published in 1982, in the French language [1]. It dealt with a (special) GLMT concerning the case of an illuminating axisymmetric incident light beam interacting with a sphere defined by its properties (d, m) , using the Bromwich formalism. The interaction is on-axis, that is to say the axis of propagation of the incident beam passes through the center of the scatterer (otherwise, the interaction is said to be off-axis). Algebraic expressions are established for scattered field amplitudes, and for scattered intensities, and specified in the far field. They introduced expansion coefficients associated with partial waves, denoted as g_n , and later on named beam shape coefficients. The obtained expressions are very close to the one of the Lorenz-Mie theory such as reported by Kerker [23] and can therefore easily be implemented in a classical Lorenz-Mie computer program, once the beam shape coefficients are calculated. The LMT itself is found to become a special case of the special GLMT.

The concept of axisymmetric light beam has been much later extensively discussed by Gouesbet [74]. An axisymmetric light beam is defined as a beam for which the component of the Poynting vector in the direction of propagation does not depend on the azimuthal angle in suitably chosen coordinates. In such coordinates, the partial wave representation of the beam is again found to be given by a special set $\{g_n\}$ of beam shape coefficients. An example of such axisymmetric light beams is a laser beam, in the mode TEM_{00} , or Gaussian beam. By contrast, a laser sheet is not an axisymmetric light beam.

In 1985, the 1982-formalism is adapted to the case of a Gaussian beam modelled as a low-order Davis beam [75] called order L beam (L for lower) [76]. This adaptation could be viewed as a specification since a Gaussian beam has been said to be a special case of axisymmetric beams. However, the axisymmetric beams used in the 1982-article were too simple to match the description of an order L Davis beam. Actually, they could match a still

simpler description of Gaussian beams called order L^- of description. Therefore, the specification to an order L Gaussian beam could also be viewed as a generalization. In the introduction of the article, we stated that we were providing a generalization of our previous contribution by modelling the Gaussian beam by field expressions more rigorous than the ones corresponding to an axisymmetric light profile. As we can see from the above discussion, this statement has now to be viewed as erroneous, due to the fact that, at that time, we did not possess a full mastering of the concept of axisymmetric light beam that became available only more than ten years later. We should have written that we were providing a generalization to axisymmetric light beams (order L Gaussian beams) more general than the axisymmetric light beams considered in 1982. Another (more significant) generalization is that further expressions are also provided for phase angles, cross-sections, efficiency factors, and radiation pressures. All these expressions involve again the special beam shape coefficients g_n .

Due to the difficulty (that we shall recurrently encounter) in providing a good enough description of Gaussian laser beams and, more generally, of any kind of shaped beams, and in providing descriptions exactly satisfying Maxwell's equations, in particular (this was not the case for the aforementioned order L and L^- descriptions of Gaussian beams), we commented at that time that the theory was not yet rigorous. This was may be too severe. It would have been interesting and more relevant to separate in the results what was general, and what was specific to the illuminating beam under study, something that was done only later, in 1988. But, nevertheless, some emphasis on Gaussian beams has been useful for future developments. Furthermore, it was also worthwhile to carefully study the properties of and the degrees of approximations involved in the orders L and L^- of description of Gaussian beams. This was done in 1985 [77].

In parallel to these developments, there was a real worry concerning the possibility of developing practical applications of the pregnant GLMT. Indeed, when we started trying to compute the beam shape coefficients g_n , it has been disappointingly discovered that these computations were too much time consuming, possibly two or three hours on the most powerful mainframe computer readily available in France at that time, for only one beam shape coefficient. Just consider now that for any realistic light scattering computation in the GLMT-framework, hundreds or thousands of such beam shape coefficients have to be calculated. The original expressions for evaluating the (special) beam shape coefficients relied on double quadratures. Eventually, another method, valid at that time for Gaussian beams, has been discovered. This method has been called the localized approximation, relying on a localized interpretation, inspired by the famous principle of localization of Van de Hulst [17]. The terminology "localized approximation" is however may be to be regretted and, certainly, is unfortunate. There is a sense in which the result obtained is an approximation because it is indeed an approximation to an ideal (unknown) Davis beam. However, there is a sense in which it

is not an approximation, namely the fact that it provides field expressions which exactly satisfy Maxwell's equations. The examination of the literature demonstrated that the word "approximation" has often been interpreted in a negative manner, some authors then proposing other beam models supposed to be rigorous, not approximations. A better terminology would have been to say from the beginning that the localized interpretation provided a localized beam model, as will be later introduced. As far as we know, all beam descriptions are models, whether they do not satisfy or do satisfy Maxwell's equations. Such issues will have to be developed more extensively in the bulk of the book.

The first archival article on the localized approximation is dated 1986, by Gréhan *et al* [78]. In this article, the localized approximation is validated by comparing the GLMT (with the illuminating beam described with a localized beam model) and a Rayleigh-Gans approximation for Gaussian illumination. Other validations are discussed too, namely comparisons with theoretical results from Tsai and Pogorzelski [62] and from Yeh *et al* [67], and also with an experimental scattering diagram under laser beam illumination obtained from a sphere in optical levitation [57]. A validation is however not a rigorous justification which will become available only nearly ten years later [79], [80]. In 1987, the first computations of beam shape coefficients by quadratures (named "first exact values") became published, with computing times ranging from 30 s to 2 hr CPU, and favourably compared with the results of the localized approximation [81]. The same year, complementary computations of beam shape coefficients g_n were published, with comparisons between "exact" values at both orders L and L^- , and the localized approximation. Also, a discussion of the physical interpretation of the localized approximation is provided and the GLMT is again used to interpret, in a more extended way than previously, an optical levitation experiment. Furthermore, a comparison between GLMT and diffraction theory in the near forward direction is discussed [82]. A similar complementary discussion, in French, is available from Maheu *et al* [83], which is the first part of a two-part article. The second part [84] provides and discusses several GLMT-based scattering diagrams, phase angle computations, efficiency factors, and collected powers, that is to say the most extensive set of results hitherto obtained with the special GLMT. It has soon be also emphasized that such computations could be successfully carried out on a micro-computer, with a maximum of 64 Ko of variables accepted by micro-computers of the PC family running under DOS 3 [85]. Note that, nowadays, LMT-computations (not GLMT-computations however) are feasible on a mobile phone! [86]. Beside quadratures and localized approximation, a third technique to evaluate the beam shape coefficients, namely by using finite series, has also been introduced. We then possessed three methods which are compared in a 1988-article [87].

It was then clear that the special GLMT was mature and ready for applications. When approaching this result, it became obvious that it was the right time to build a final general version of the theory. A first version, for

arbitrary location of the scatterer, is discussed by Gouesbet *et al* in 1988 [88], with a strong emphasis, including in the title, on Gaussian beams. However, as stated in the conclusion, other beam descriptions could be used instead of a Gaussian beam, without changing the method used. In particular, this reference introduced new expressions and new beam shape coefficients which do not depend on the specific illuminated beam considered. These new beam shape coefficients, generalizing the special beam shape coefficients g_n , are denoted as $g_{n,TM}^m$ and $g_{n,TE}^m$. Changing the beam description just implies to evaluate these general beam shape coefficients in a new way, adapted to the new description (or more generally to the new beam under study, e.g. laser sheets). A second more extended version, that we consider as the "pivot" article of GLMT, has also been published in 1988 [2]. Again, may be unfortunately, a strong emphasis is put on Gaussian beams, including in the title of the article, but the fact that GLMT works for arbitrary-shaped beams is made explicit in a companion article published in the same year [89], devoted to GLMT for arbitrary location of a scatterer in an arbitrary profile.

We may now pursue our brief history of GLMT by relying on a few review articles published on the topic, allowing one to follow subsequent developments in a concise way. The first review article was published in 1991 and, most essentially, presented the GLMT formalism under a single roof [90]. The next review article was published in 1994 [91]. The general formulation was required and it is once more pointed out that this formulation is insensitive to the nature of the incident beam. More important, genuine applications to optical particle characterization, in agreement with our original motivation, could be discussed, namely phase Doppler anemometry and trajectory ambiguity effects. Other miscellaneous applications concerned scattering responses and extinction cross sections, diffraction theory, optical levitation and radiation pressure. The third review article, in 2000 (with about 350 references) reported on new theoretical advances, in particular by discussing infinitely long cylinders and other shapes, and extended applications : radiation pressure, rainbows, imaging, morphology-dependent resonances, phase-Doppler instruments, etc. It also provided recommendations for future research [92]. See also Gouesbet [93] and Gouesbet *et al* [94].

Now, something special happened in 2008, namely that it was the hundredth anniversary of the famous Gustav Mie's article. This has been commemorated in several places (GAeF conference 2008 on "Light Scattering : Mie and More-commemorating 100 years Mie's 1908 publications", 3rd-4th July, Karlsruhe, Germany [95]; International Radiation Symposium IRS 2008, 3rd-8th August, Foz do Iguaçu, Brazil [96]; Eleventh Conference on Electromagnetic and Light Scattering, 7th-12th September 2008, Hatfield, UK [97]; Mie theory 1908-2008, Present Developments and Interdisciplinary Aspects of Light Scattering, 15th-17th September, Universität Halle-Wittenberg). It has been an opportunity for two more review articles [98], [99]. In particular, the second one (with again about 350 references) exhibited the fact that the use of generalized Lorenz-Mie theories and associated ingredients was more

and more widespread, confirming the need for a book like the present one. Let us also mention a review of elastic light scattering theories, available from Wriedt [100], and the existence of a *scattering information portal* for the light scattering community discussed by Wriedt and Hellmers [101].

Furthermore, the series of published articles suffered from three main shortcomings. The first one is that, as usual in research, the development of our ideas did not follow a linear line. The reader wanting to fully exploit our study and check our derivations would then be condemned to the burden of reorganizing the published material. The second shortcoming is that many details have been omitted, the usual (unfortunately justified) prayer of referees and editors being to ask for drastic cuts. To go from relation n to relation $(n + 1)$, 20 intermediary pages of computations must sometimes be reproduced by the reader who is therefore left with a skeleton from which living flesh has been carefully removed to produce an article written in an objective, concise, modern scientific style. Finally, in practice, program sources are not being published in the archival literature. Scientists playing tennis usually ask to the authors to get the ball i.e. program sources, but the ones playing golf and wanting to move the ball alone must also reproduce non trivial computer programs.

These three shortcomings are essentially avoided in this book. The material is reorganized in a linear and comprehensive way. We take the reader by the hand and guide him in the forest, following a civilized track for berry pickers. People just wanting to get numerical results may have a bird's eye view on the formulation and may directly go to the computer programs to use them. Hopefully, this book might then be useful to and appreciated by both tennismen and golf players.

Although this book is dedicated to electromagnetism, there has been a few moves toward quantum mechanics, which are now briefly mentioned. The structure of quantum arbitrary shaped beams has been examined [102], [103]. Quantum cross-sections under quantum arbitrary shaped beam illumination are discussed for both elastic [104] and inelastic [105] scattering. Cross-sectional analogies between (vectorial) electromagnetic scattering and (scalar) quantum scattering have also been established, again both for elastic and inelastic cases, under plane wave and under quantum arbitrary shaped beam illuminations [106], [107], [108]. Also, a generalized optical theorem for non plane wave scattering in quantum mechanics has been established [109].

The book is organized as follows:

Chapter I provides a background in Maxwell's electromagnetism (in free space and in matter) and discusses Maxwell's equations. The content of this chapter is supposed to be sufficient to attack the rest of the book, but it is not meant to provide a detailed introduction to electromagnetism. Preferably, the reader should already possess some kind of basic knowledge to enter this book. Chapter I rather intends to recall what is necessary to proceed further and to introduce notations, as well as the basic language to be used.

Chapter II introduces the method of Bromwich scalar potentials which has been originally used to solve Maxwell's equations for the main case under study, that is to say to build the GLMT. Relationships between this method and other equivalent possible methods (such as the use of vector spherical wave functions) are also discussed.

Chapter III uses the material introduced in the first two chapters to describe the GLMT for arbitrary location of the scattering sphere in an arbitrary incident beam, that is to say without referring to any specific kind of illuminating beam. This theory introduces two sets of beam shape coefficients denoted as $g_{n,TM}^m$ and $g_{n,TE}^m$ which describe the illuminating beam and are recurrently used in GLMT-expressions. The expressions for the quadrature methods to evaluate these beam shape coefficients are discussed (although they are lengthy and costly to perform). Other formulations to deal with arbitrary shaped beam scattering are briefly considered. Other generalized Lorenz-Mie theories (i.e. for different shapes of the scatterer) are also introduced.

Chapter IV discusses specific beams, with a very strong emphasis on Gaussian beams.

Chapter V establishes that beam shape coefficients may also be computed by using a finite series method which is computationally more efficient than quadratures. The introduction of these finite series provided a first significant step to speed-up GLMT-computations.

Chapter VI is devoted to the special case when the center of the scattering particle is located on-axis in an axisymmetric beam, such as a Gaussian beam (or a plane wave!), leading to dramatic simplifications. In particular, the double set $\{g_{n,TM}^m, g_{n,TE}^m\}$ of beam shape coefficients reduces to a single set $\{g_n\}$ of special beam shape coefficients. The similarity between LMT and GLMT in this case is striking, and we easily recover LMT from GLMT as another more special case. Also, a significant interest of this case is that the formulation becomes very similar to the one of the classical LMT, with the result that computer programs for LMT can readily be adapted to the special GLMT, once the beam shape coefficients are evaluated.

Chapter VII discusses a very beautiful method to evaluate beam shape coefficients (g_n , $g_{n,TM}^m$, and $g_{n,TE}^m$), our favourite one indeed. It is actually the fastest one and provides many physical insights on the meaning of the beam shape coefficients. It has been called (may be unfortunately) the localized approximation, relying on a localized interpretation. It generates localized beam models which exactly satisfy Maxwell's equations.

Chapter VIII discusses concisely but fairly exhaustively the applications of GLMTs.

The bulks of the chapters provide basic knowledge, ordered in, we hope, a rather logical order. Most of these bulks are accompanied by complements. The aim of these complements is to address the reader to more technical or complementary matters, and to an exhaustive literature. Although they provide valuable information, allowing the reader to deepen the topic if required, their reading can be omitted at first without provoking any havoc in the understanding of the subsequent material dealt with. These complements are written in a concise way. Owing to this fact, they allow the reader to gain contact with all the material available at the present time (except for possible unfortunate omissions for which we apologize), without having to manage with an over-sized book. Chapter VIII is written in the same style than the complements.

To prepare the complements and Chapter VIII, we relied on ISIweb of knowledge, and made a list by extracting the articles citing the Rouen works on GLMTs. A significant number of them (but not all of them) are cited in this book. Some articles have been removed from the citing list when they are not enough relevant to the aim of this book (this should not be considered as a negative appraisal however). Conversely, some articles which do not pertain to the list have been used when they are found to be useful for a better understanding of the exposition and of the chronology of events. The corresponding cited articles may be arranged in three families. A first family corresponds to articles which explicitly use or rely on GLMTs. In a second family, GLMTs have not been used but could have been used. This is the family of GLMT-izable articles. A third family concerns citing articles which are not strictly relevant to GLMTs but are relevant to the more extended field of light scattering as a whole. They may give the reader a flavour of the environment in which GLMTs have to move. Furthermore, as a consequence of the above strategy for complement-like issues, the corresponding cited articles are not necessarily the first articles that were published in each individual topic. Rather, they may be articles pertaining to the citing list that may build on other earlier studies. These earlier studies may be reached from the reference lists of the cited articles. This discussion defines the sense in which the complements and Chapter VIII are exhaustive.

Appendices expose some technicalities of secondary significance. However, one of them should attract the particular attention of many readers, namely it contains a list of computer programs provided in a website connected to the book. This website also contains movies showing the interaction between some scatterers and ultra-short pulses.