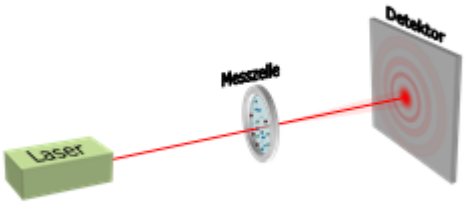
# Laser Scattering – A brief Introduction

**The Milky Way: Luminous Gas and dark Dust**

In the twenties of the last century, the Swiss astronomer R.J. Tümpler discovered that distant star clusters appeared darker than was to be expected from their distance. He concluded that part of the starlight gets lost on its way to our Earth. A few years later, the American astronomer E.P. Hubble observed that the average number of visible galaxies in the direction towards the center of our Milky Way in the constellation Sagittarius is much smaller than when looking towards the Big Dipper, for example. In addition to brightly shining gas clouds, predominantly consisting of hydrogen, one also finds numerous dark regions in the so-called galactic plane, which almost completely swallow the light coming from objects behind them.
The interstellar dust was discovered.
Interstellar dust consists of mostly very small particles - their typical diameter is between 0.1 and 1 µm - that scatter and absorb starlight. Since these particles cannot be studied by alternative methods such as electron microscopy, the application of light scattering theory has been and still is of great importance to astronomers in the study of interstellar or even interplanetary dust.
In the laboratory, for earthly applications as it were, the conditions are somewhat simpler. Or perhaps it would be better to say: The challenges are different. The optical design of the overall system can of course be adapted to the requirements here, and one usually knows more about the sample material under investigation than in the case of space particles. Starlight with its broad wavelength spectrum can be replaced by monochromatic laser light, and the chemical composition of the sample material is often well known. On the other hand, new difficulties arise, especially in the suitable preparation of the particle collective to be measured. But first things first. Let us start with the basic design of a suitable device for measuring particle size distribution by laser scattering.

**Basic Design of a Laser Particle Sizer**

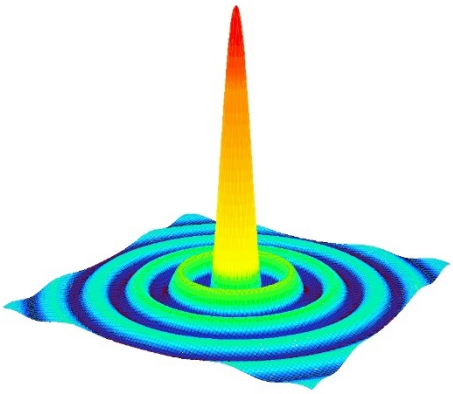
In principle, the setup is always the same: A light beam, usually delivered by a laser, passes through the sample to be measured and behind this the intensity distribution generated by diffraction or scattering is recorded by a detector. Already at this point it should be mentioned that the particle collective to be measured should be provided in a sufficient dilution and should not form any agglomerates. The measured intensity distribution then shows a system of numerous more or less concentric rings, whose spacing correlates with the particle size.
Large particles produce closely spaced rings, small particles more widely separated rings. The particle size can be calculated from the distance between these individual rings.



**Scattering Theories**

The theories used for this purpose are either the **Fraunhofer theory** (also called Fraunhofer approximation) or the **Mie theory**.

In the **Fraunhofer theory**, the measured intensity distribution is described exclusively by the so-called diffraction of the laser light at the particles (this is also the reason for the somewhat simplifying common use of the term "laser diffraction").



Diffraction is a phenomenon that always occurs when a wave interacts with an obstacle such as a particle. By superposition of different parts of the wave front disturbed by the particle (interference), a characteristic diffraction pattern occurs behind the particle, the exact distribution of which is described by Fraunhofer diffraction. The figure on the right is a graphical representation of the scattering amplitude of a spherical particle. Plotted vertically (and additionally color coded) is the amplitude of the scattered light (i.e. the intensity[1]). The distance from the center of the image indicates the scattering angle, i.e. in the center of the image the scattering angle is zero (no scattering) and towards the edges the angle increases. You can recognize clearly the central diffraction maximum for only very small scattering angles, which shows the highest intensity of scattered light. Towards larger scattering angles - and thus to larger distances from the detector center of a corresponding measuring instrument - then follow alternating dark and bright rings, whose distance, as already mentioned above, is directly related to the particle diameter: The narrower the rings the larger the particles and vice versa.

The great advantage of the Fraunhofer theory is that no optical parameters of the sample material need to be known. However, it must be pointed out that initially a spherical geometry is assumed.

In many cases, the use of Fraunhofer theory is sufficient for particle sizing. But for non-transparent and especially for small particles, one cannot stick to diffraction alone. The limits of Fraunhofer theory are reached when the particle size approaches the wavelength of the laser light used.

**Mie Scattering**

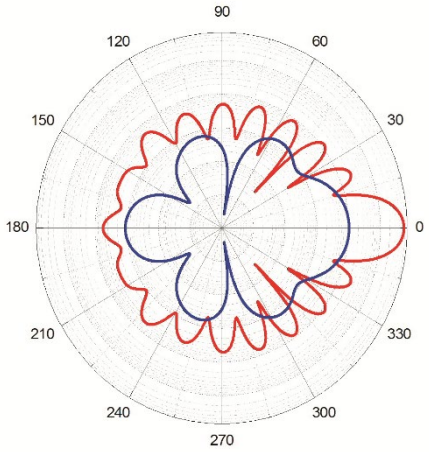
For particle diameters on the order of the wavelength of light and below, Mie theory then comes into play. The Mie theory is the complete solution of Maxwell's equations for the scattering of electromagnetic waves by spherical particles. Now what does this mean? Well, you can imagine that the electromagnetic light wave couples to the atoms and molecules in a particle, so to speak, and causes them to oscillate. These oscillations then in turn generate electromagnetic light waves of the same wavelength (we are talking only about elastic scattering here), which are radiated in all possible directions. Superposition of the individual waves from the different areas of the particle then leads to the formation of a characteristic intensity distribution, which, unlike in Fraunhofer diffraction, can be observed not only in the forward direction but also at scattering angles greater than ninety degrees.

Starting from Maxwell's equations, which describe the propagation of electromagnetic waves in general, Gustav Mie investigated the effects of light scattering in colloidal metal solutions, especially the scattering of light by fine gold particles, at the beginning of the 20$^{th}$ century and was one of the first to develop a complete theory, which was later named after him.

[1] Strictly speaking, the intensity is of course the square of the amplitude

**The Polar Diagram**

With the help of a polar diagram, the angle-dependent intensity distribution of the scattered light can be displayed very clearly. The diagram is read as follows: The numbers at the outer edge of the diagram indicate the scattering angle. The distance between the centre of the diagram and the coloured distribution curve then indicates the intensity of the light scattered in that direction. It should be noted that the radial intensity axis is logarithmic. Comparing the blue line with the red line, we see that for larger particles - red - the intensity for small scattering angles - 0° to about 15° - is about one hundred times stronger than in the backward direction at near 180°. For the blue curve, this difference is already much smaller. From this we learn that for very small particle diameters the intensity of the backward scattered light becomes important.




Scattering intensity of red laser light scattered once by gold particles with a diameter of 1.5 µm - red line - and once by gold particles with a diameter of 0.5 µm - blue line.

Similar to the procedure for Fraunhofer diffraction, the intensity distribution can now be used again to calculate the particle size. The difficulty with the calculation according to Mie is, however, that here, in contrast to the Fraunhofer theory, the knowledge of material constants of the investigated system is necessary. More specifically, the real and imaginary parts of the complex refractive index, i.e. refractive index and absorption coefficients (often referred to collectively as Mie parameters), are required for both the sample material and the dispersion medium used, and this for each wavelength of the light used - or, in the case of several different light wavelengths, even all indices for all wavelengths. Despite extensive databases, which at least provide the refractive index of numerous materials, for many sample systems as found in daily laboratory practice, the corresponding parameter sets are not available and must first be determined by time-consuming investigations. A short word of warning is allowed here: Repeatedly one reads about the fact that with the use of measurements of the particle size distribution by laser scattering the Mie parameters can be determined. Unfortunately, this is not possible in such a simple way - which is not surprising, since it is precisely these parameters that are needed to calculate the particle size. Nevertheless, there are possibilities to gain some knowledge about the Mie parameters. But this will be briefly discussed later.

**The Technology: Optical Setup**

Now that the basic theories of light scattering have been described, at least in brief, the more detailed design of the optical setup of a laser particle sizer will be described. The basic arrangement already outlined above can be realized with two different concepts. In addition to the components mentioned (laser - measuring cell - detector), a collecting lens (Fourier lens) must be integrated into the beam path to focus the scattered light onto the detector. The arrangement of this Fourier lens makes the decisive difference between the so-called **conventional design** and the **inverse Fourier** setup.

**Conventional Setup**

Here, a sufficiently wide, parallel laser beam is generated, into which the measuring cell with the scattering particles is then introduced. The Fourier lens is positioned between the measuring cell and the detector. Since the focal length of the Fourier lens determines the measuring range in this arrangement, the lens must be exchanged in order to obtain a different range. This must be adjusted with high accuracy, since very small angles are measured, especially with larger particles, and a tilt of the Fourier lens has a direct

influence on the measurement result. Another disadvantage of this arrangement is the limited possibility to measure large scattering angles. And these large scattering angles are, as we know, necessary for the measurement of very small particles.

**The inverse Fourier Design**

In the 1980s, FRITSCH GmbH therefore introduced the inverse Fourier design as an alternative with the first model of the ANALYSETTE 22 series. In contrast to the conventional design, here the Fourier lens is located in front of the measuring cell, so that this is not illuminated by a parallel, but by a convergent laser beam. The scattered light is thus focused directly on the detector without any further optical elements. Despite initial scepticism from many manufacturers, the basic principle of the Inverse Fourier setup has now proven itself in many devices and is also described alongside the conventional setup in ISO standard 13320, which is relevant for laser diffraction. The advantage lies in the comparatively simple setup, in which no parts have to be moved to cover the entire measurement range.

**The Mathematics: Calculation of the Particle Size Distribution**

As already mentioned, almost all samples relevant in practical applications do not consist of particles of identical diameter. Rather, one finds a quasi continuum of particle size and thus a superposition of all respective scattered light distributions. This means that, compared with only one particle diameter, a less structured measurement signal is obtained, from which the individual particle diameters and their relative proportions in the sample are now to be calculated.

For this purpose, the entire measurement range is first divided into individual intervals and then a system of equations is set up which describes the scattered light intensity for each of the detector elements present for each particle size interval. The solution of this equation system then provides the respective relative proportions, i.e. the particle size distribution sought. This is, by the way, also the reason why this particle sizing technique has only been commercially available since the 1980s. Before that, the computing power required to solve the equation systems was not available at a reasonable price.

Unfortunately, the equation systems to be solved are unstable. Unstable means that even the slightest changes in the input values - the measurement data - can lead to drastic changes in the results. This behaviour is not device-specific, but is inherent in the nature of the method. However, there are different mathematical methods to control these instabilities so that stable, reproducible and accurate results can still be achieved. Care must be taken that the stabilization of the equation systems is not too strong, since this results in an increased smoothing of the result and any details of the distribution that may be present are lost.

As discussed above, when using Mie theory, the choice of optical parameters sometimes have a significant effect on the solution of the equation system, i.e. on the result. Here then arises the aforementioned possibility of at least narrowing the choice of Mie parameters. The strategy is as follows: With a best possible chosen set of Mie parameters one determines the particle size distribution from the measured intensity distribution of the scattered light. With this size distribution, one now goes the opposite way again: one determines the expected angle-dependent intensity distribution of the scattered light using the chosen Mie parameters. This process is comparatively simple and is called simulation. Then one compares the calculated (simulated) intensity distribution with the actual measured distribution. How well the simulated and the actually measured intensity distribution match gives an indication of the reliability of the result and thus also of the correctness of the chosen Mie parameters. You can now change the Mie parameters and repeat the procedure. In practical applications, one will generally find that the agreement between the simulated and the measured intensity distribution is not perfect, since numerous factors influence the intensity distribution. Besides always existing disturbances during the measurement, these are of course deviations from the ideal spherical shape and inhomogeneities of the particles.

Thus, it cannot be assumed that one can determine the Mie parameters just by comparing the simulation with the measurement. Nevertheless, the procedure is a helpful means for the selection of suitable parameters and can be carried out elegantly with the ANALYSETTER 22 software, for example.

**Results**

Laser scattering determines the relative volume distribution of the measured sample. This means that the result of a measurement using laser scattering tells you, for example, what percentage of the total sample volume is contained in particles smaller than a certain particle size. This figure is usually called **$Q_3(x)$.**

Or you can find out what percentage of the total sample volume is contained in particles that lie within a certain size interval. This key figure is then called **$dQ_3(x)$.**

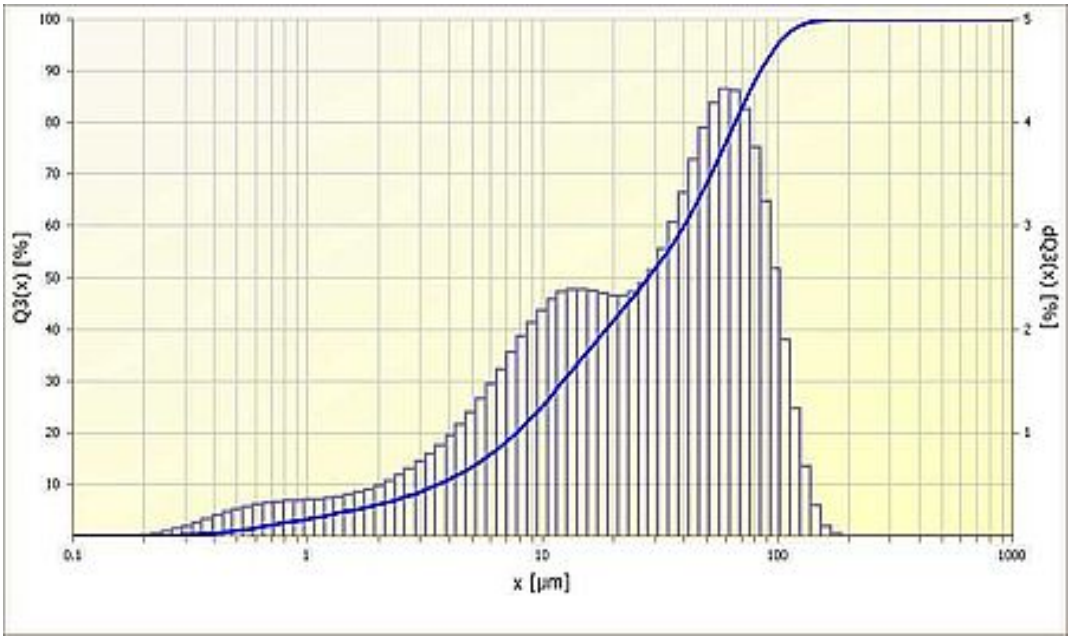



**Illustration:** Particle size distribution of fly ash measured with an ANALYSETTE 22. The solid line is the so-called cumulative curve $Q_3(x)$, the bars represent the values of $dQ_3(x)$.

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