Chapter 1

Introduction.

Elastic light scattering (LS) has been extensively used to study samples showing a non uniform refraction index on lengthscales from a fraction of a micrometer to a fraction of a millimeter. Basically, a wide laser beam is sent through the sample, and the light scattered at any angle is measured by a detector in the far field. Many phenomena have been studied with this technique; among them, thermodynamical fluctuations of concentration in solutions, of temperature, of pressure, convective instabilities and turbulence, colloids and colloidal gels, systems showing critical opalescence.

The measure of the intensity of the beams scattered by a target is a well known way for analysing physical properties of a sample, and is used in many fields of physics: examples are X ray diffraction in crystallographic analysis and particle scattering by nuclei in the Rutherford experiment. In these examples, as in light scattering, the intensity of the scattered beams gives informations on the square modulus of the Fourier transform of a quantity of interest of the sample, the so called power spectrum, evaluated in the transferred wavevector. In the case of light scattering, the quantity of interest is the refraction index. Since refraction index shows variations due to concentration, temperature and pressure fluctuations, all these quantities can be investigated using light scattering.

In principle, measuring the intensity of the light scattered at an arbitrary small angle allows to obtain informations on arbitrary long wavelength Fourier components of the sample. In facts, the measurement of the intensity of the scattered light becomes more and more difficult as the scattering angle becomes small, mainly due to the stray light, that is the light scattered by the imperfections of the optical system, which is mainly scattered at small angles. Thus light scattering cannot give informations on the features of a sample, if their associated wavelenght is longer than a given value. On the other hand, image forming techniques, such as Schlieren, dark field, phase contrast microscopy, have no limitations on the size of the features they can observe. The limitation of many image forming techniques is the difficulty to quantitatively relate the observed images to the physical properties, mainly when dealing with three dimensional samples.

In the present work, we describe three new techniques, which allow to measure the scattering intensities, overcoming the difficulties associated to small angles. The first of these techniques, the hOmoyne Near Field Speckles (ONFS), has been presented very recentely [1]; in the present work, we show new results, obtained with a slightly improved optical setup [2]. The other techniques, the hEterodyne Near Field Speckles (ENFS) and the Schlieren-like Near Field Speckles (SNFS) are improvements based on that. The first has been recently patented by us [3, 4]; the second is presented here for the first time. Moreover, in Chapt. 3, we present for the first time a mathematical derivation of the working formulas for the three techniques.

Basically, the experimental setup consists in a wide laser beam passing through the sample; a lens forms an image of a plane at a given distance from the cell on a CCD sensor. The image, in the near field, shows speckles, since it is formed by the stochastical interference of the light coming from a random sample: the electric field has a gaussian probability distribution. We will show that, under suitable conditions, the correlation function of such a field closely mirrors the correlation function of the investigated sample; moreover, in general, from the correlation function of the speckle field we can calculate the scattered intensity.

The lens that forms the image on the CCD focuses the transmitted beam around a given point. In ONFS, a beam stop is placed in that point, in order to dispose of the transmitted beam; in SNFS a blade stops half transmitted beam, along with one half of the scattered light, like in Schlieren technique; in ENFS no opaque element is introduced in the optical system. In ONFS, the CCD sees the speckles given by the interference of the scattered beams with themselves: ONFS is an homodyne technique . In ENFS and SNFS, the speckle field is heterodyned with the much more intense transmitted beam, that acts as a reference beam: the measured intensity is linear in the speckle electric field. We acquire a set of images, from one of the three techniques, by using a CCD camera, connected to a frame grabber; the images are then elaborated by a PC, to obtain $I(\vec{q})$, the scattered intensity as a function of the transferred wavevector, the same information obtained by LS. For each technique, we developed algorithms which allow to evaluate the scattered intensities, including the corrections for the stray light and for the shot and read noise of the CCD camera. The algorithms are described in Chapt. 5 and 6.

We used ONFS and ENFS to measure the scattered intensity of some colloids, and we compared the results with those made by a state-of-the-art classical Small Angle Light Scattering (SALS) device. The agreement is very good, notwithstanding a much simpler and stable layout. In Chapt. 7 we compare ONFS, ENFS and SALS measurements, and discuss the main sources of errors. Scattering intensities measured with ENFS show a better quality; we used this technique to evaluate the diameter distribution of some known colloids, by using an inversion algorithm based on Mie theory. The results are presented in Chapt. 8; this shows that ENFS is a simple and powerful alternative to SALS, suited for industrial applications like particle sizing. Moreover, SNFS has been used to evaluate the power spectrum of non-equilibrium fluctuations in a free diffusion process, thus showing that such techniques have interesting applications in fundamental physics; results are shown in Chapt. 9.