Chapter 6

ENFS and SNFS data processing.

In this chapter we describe the algorithm used to process ENFS and SNFS data, acquired by a CCD, in order to evaluate of the scattered intensity. From a set of images, taken at different times, we are able to subtract the stray light, point by point: this is a noteworthy feature of the heterodyne techniques. The algorithm we describe is similar to the one which has been applied to shadowgraph, and should work for every heterodyne technique.

The algorithm has been applied to SNFS, and the results are shown in Chapter 9; in this chapter, all examples refers to colloid measurements made with ENFS.

6.1 Subtraction of the stray light

As in every heterodyne technique, we measure the overall intensity $I\left(\vec{x}\right)$, generated by the interference of an object field $\delta E\left(\vec{x}\right)$ with a more intense reference beam with amplitude E_0. In our case, the object field is generated by the scattered beams, and the reference beam is the transmitted one. We measure the heterodyne signal $i\left(\vec{x}\right)$:

$$i\left(\vec{x}\right) = \frac{I\left(\vec{x}\right) - I_0}{I_0}, \tag{6.1}$$

where I_0 is the mean intensity. At first order in $\delta E/E_0$, Eq. (3.22) holds:

$$i\left(\vec{x}\right) = 2\frac{\Re\left[\delta E\left(\vec{x}\right)\right]}{E_0}, \tag{6.2}$$

where we have assumed that E_0 is real. Equation (3.72) states that, under the conditions in which NFS works, the power spectrum of $i\left(\vec{x}\right)$ is the power spectrum of the electric field, the quantity we must measure in order to evaluate the scattered intensities.

We developed an algorithm to subtract the contribution of the stray light, directly on each image, point by point. This is a noteworthy feature of the heterodyne techniques, since in dynamic light scattering and in ONFS the subtraction is possible only on the scattered intensity or on the correlation function, averages of square values. The scattered field can be decomposed into $E_{SL}(\vec{x})$, the stray light field, and $\delta E(\vec{x})$, the field of the light scattered by the sample. Both $E_{SL}(\vec{x})$ and $\delta E(\vec{x})$ are much less intense than E_0, the reference field. At the first order, the resulting intensity is:

$$I(\vec{x}) = E_0^2 + 2E_0\Re\left[E_{SL}(\vec{x})\right] + 2E_0\Re\left[\delta E(\vec{x})\right] \tag{6.3}$$

In many cases, $\delta E(\vec{x})$ fluctuates in time and is correlated only on finite delays. On the contrary, stray light comes mainly from hard surfaces, and does not change as times go on. This is the case of the samples we studied. The spatial average of a scattered field is alwais wanishing; this property, along with the absence of correlation on different images, says that the average over many images of $\delta E(\vec{x})$ vanishes.

In order to separate the contribution of the stray light, we average $I(\vec{x})$ over many different images. Since the phase of $\delta E(\vec{x})$ is random, its mean vanishes:

$$\{I(\vec{x})\} = E_0^2 + 2E_0\Re\left[E_{SL}(\vec{x})\right]. \tag{6.4}$$

We use the symbol $\{\cdot\}$ for the mean over many samples, and the symbol $\langle\cdot\rangle$ for the mean over $\vec{x}$. The fluctuation $I(\vec{x}) - \{I(\vec{x})\}$ does not depend on $E_{SL}(\vec{x})$:

$$I(\vec{x}) - \{I(\vec{x})\} = 2E_0\Re\left[\delta E(\vec{x})\right]. \tag{6.5}$$

Because of the conservation of the total intensity during the scattering process, by averaging $\{I(\vec{x})\}$ over the whole plane, we obtain I_0:

$$\langle\{I(\vec{x})\}\rangle = E_0^2 = I_0. \tag{6.6}$$

We can now evaluate the heterodyne signal $i(\vec{x})$, subtracting the the stray light contribution:

$$i(\vec{x}) = \frac{I(\vec{x}) - \{I(\vec{x})\}}{\langle\{I(\vec{x})\}\rangle}. \tag{6.7}$$

6.2 Correction for finite samples.

The quantity $\{I(\vec{x})\}$ should ideally be evaluated by averaging infinite images. We obtain a good evaluation of it by averaging a great number N of images $I_n(\vec{x})$, typically one hundred:

$$\bar{I}(\vec{x}) = \frac{1}{N}\sum I_n(\vec{x}) \approx \{I(\vec{x})\} \tag{6.8}$$

From this evaluation, we obtain $i(\vec{x})$:

$$i_n(\vec{x}) = \frac{I_n(\vec{x}) - \bar{I}(\vec{x})}{\langle\bar{I}\rangle} \tag{6.9}$$

The average value $\bar{I}\left(\vec{x}\right)$, evaluated over a given number of images, is sistematically different from the true mean value, in the direction that reduces the evaluation of the root mean square displacement from the mean. This problem is analogous to the one that leads to the so called Bessel correction for the evaluation of the variance σ of a stochastic variable, from the knowledge of a finite number of stochastic values.

We evaluate the correlation function of $i_n\left(\vec{x}\right)$ for each n, then we average them, thus obtaining $C_i\left(\Delta\vec{x}\right)$. Now we want to evaluate $\{C_i\left(\Delta\vec{x}\right)\}$, that is the mean value over infinite samples, in order to correct systematic errors:

$$\{C_i\left(\Delta\vec{x}\right)\} = \frac{1}{\left\langle\bar{I}\right\rangle^2}\left\{\frac{1}{N}\sum_{n=0}^{N}\left\langle\left[I_n\left(\vec{x}\right) - \frac{1}{N}\sum_{m=0}^{N} I_m\left(\vec{x}\right)\right]\left[I_n\left(\vec{x}+\Delta\vec{x}\right) - \frac{1}{N}\sum_{m=0}^{N} I_m\left(\vec{x}+\Delta\vec{x}\right)\right]\right\rangle\right\} \tag{6.10}$$

The symbol $\langle\cdot\rangle$ means the average over $\vec{x}$. We can write$\bar{I}\left(\vec{x}\right) + \delta I\left(\vec{x}\right)$ instead of $I_n\left(\vec{x}\right)$:

$$\{C_i\left(\Delta\vec{x}\right)\} = \frac{1}{\left\langle\bar{I}\right\rangle^2}\left\{\frac{1}{N}\sum_{n=0}^{N}\left\langle\left[\delta I_n\left(\vec{x}\right) - \frac{1}{N}\sum_{m=0}^{N} \delta I_m\left(\vec{x}\right)\right]\left[\delta I_n\left(\vec{x}+\Delta\vec{x}\right) - \frac{1}{N}\sum_{m=0}^{N} \delta I_m\left(\vec{x}+\Delta\vec{x}\right)\right]\right\rangle\right\} \tag{6.11}$$

Evaluating the products:

$$\{C_i\left(\Delta\vec{x}\right)\} = \frac{1}{\left\langle\bar{I}\right\rangle^2}\frac{1}{N}\sum_{n=0}^{N}\{\langle\delta I_n\left(\vec{x}\right)\delta I_n\left(\vec{x}+\Delta\vec{x}\right)\rangle\} - \tag{6.12}$$

$$\frac{1}{\left\langle\bar{I}\right\rangle^2}\frac{1}{N^2}\sum_{n,m=0}^{N}\{\langle\delta I_n\left(\vec{x}\right)\delta I_m\left(\vec{x}+\Delta\vec{x}\right)\rangle\} - \frac{1}{\left\langle\bar{I}\right\rangle^2}\frac{1}{N^2}\sum_{n,m=0}^{N}\{\langle\delta I_m\left(\vec{x}\right)\delta I_n\left(\vec{x}+\Delta\vec{x}\right)\rangle\} + \tag{6.13}$$

$$\frac{1}{\left\langle\bar{I}\right\rangle^2}\frac{1}{N^3}\sum_{n,m,l=0}^{N}\{\langle\delta I_m\left(\vec{x}\right)\delta I_l\left(\vec{x}+\Delta\vec{x}\right)\rangle\} \tag{6.14}$$

Since $\bar{\delta I} = 0$:

$$\{C_i\left(\Delta\vec{x}\right)\} = \frac{N-1}{N}\frac{1}{\left\langle\bar{I}\right\rangle^2}\{\langle\delta I\left(\vec{x}\right)\delta I\left(\vec{x}+\Delta\vec{x}\right)\rangle\} \tag{6.15}$$

Now we can use Eq. (6.5):

$$\{C_i\left(\Delta\vec{x}\right)\} = \frac{N-1}{N}C_E\left(\Delta\vec{x}\right) \tag{6.16}$$

The correlation function evaluated on N samples is proportional to the correlation function evaluated for $N \to \infty$. The poportionality constant is the same of the well known Bessel correction.

6.3 ENFS data processing.

Once the experimental apparatus has been built, as described in Chapter 4, in the absence of the sample, the CCD should be illuminated in a quite uniform

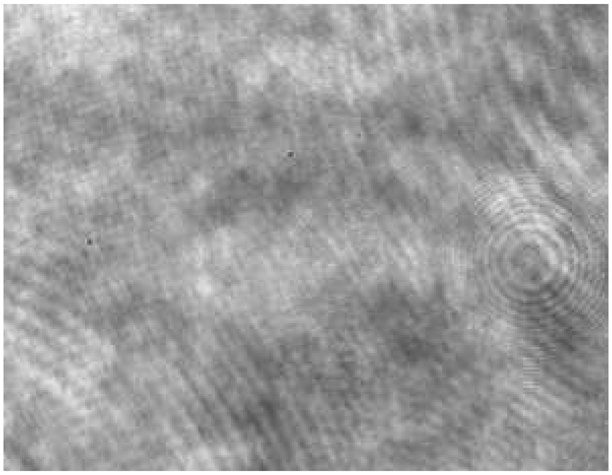

Figure 6.1: Background image.

way. As a matter of fact, the illumination is never completely uniform, primarily because of the interference of the main beam with stray light. A typical image is shown in Fig. 6.1. We can easily see some sets of concentric circles, each due to reflections inside a lens, along with speckle patterns properly due to stray light.

When the the sample is placed in the right position, we acquire about one hundred images for each measurement. The electronic shutter of the CCD and its interlacement time must be so short that no evident evolution of the system happens during the exposure: for the samples we studied, an interlacement delay of 1/25s is sufficient. Moreover, different images must be completely uncorrelated. For a 10μm colloid, images must be grabbed at intervals longer that one minute, if only brownian movements are the source of decorrelation, while for the non equilibrium fluctuations we studied the images can be taken at intervals of 1s. In figure 6.2 and 6.3 we show two typical ENFS images, generated by the interference of the main beam with the light scattered by colloids of 5.2μm and 10.0μm. The images show a mean intensity, modulated by the interference with the speckle pattern. We can notice the different typical size of the speckles. The set of concentric circles can be seen yet: the stray light will be removed with the following step.

Once the images $I_n\left(\vec{x}\right)$ have been acquired, they are averaged, in order to evaluate $\bar{I}\left(\vec{x}\right)$ and $\langle\bar{I}\rangle$. By using Eq. (6.7), we evaluate $i\left(\vec{x}\right)$, the heterodyne signal. Figures 6.4 and 6.5 show the heterodyne signal: since $i\left(\vec{x}\right)$ is negative, for some points, a constant intensity has been added. The images thus simply represent the ENFS images, cleaned from stray light and optical imprefections.

The heterodyne signal of each image is then elaborated in order to obtain its

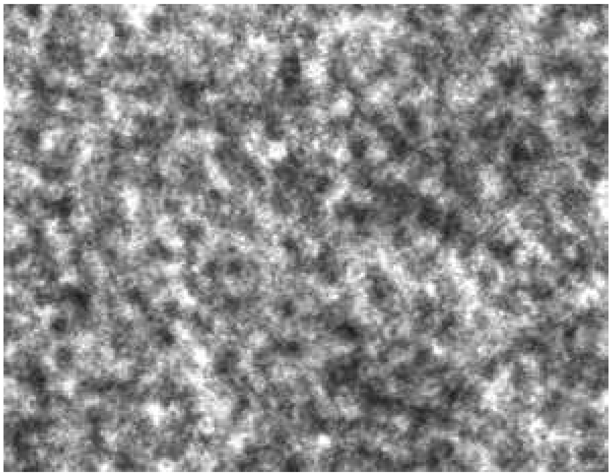

Figure 6.2: ENFS image of a 10.0μm colloid.

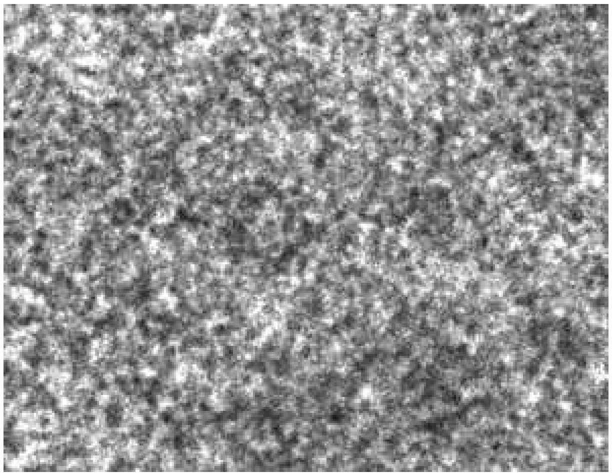

Figure 6.3: ENFS image of a 5.2μm colloid.

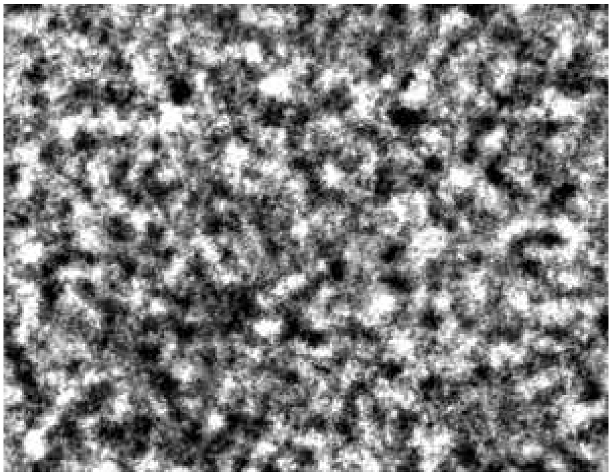

Figure 6.4: ENFS signal of a 10.0μm colloid.

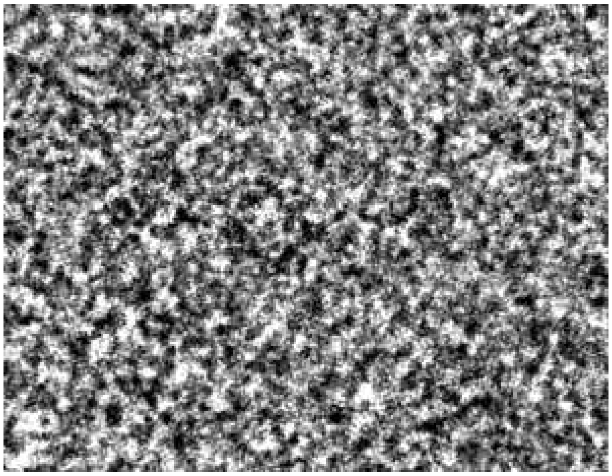

Figure 6.5: ENFS signal of a 5.2μm colloid.

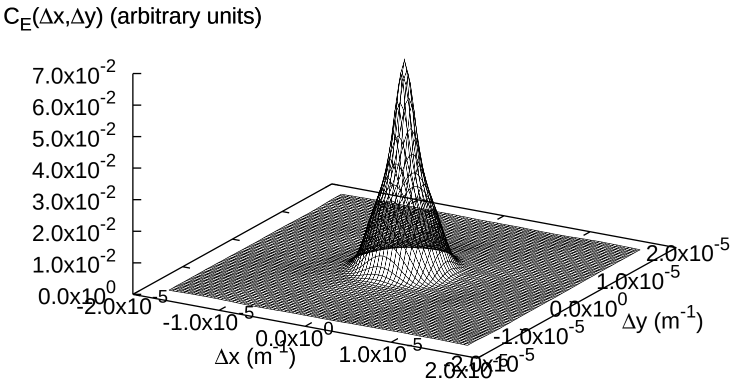

Figure 6.6: ENFS measurement of the field correlation function, for a 5.2μm colloid.

power spectrum. Simple Fourier transforming of the signal would be uncorrect, due to border effects. First of all, we evaluate the correlation function. This operation is quite fast, since we can use a Fast Fourier Transform (FFT) algorithm. An FFT algorithm allows to evaluate the Fourier tranform of an $M \times N$ matrix, with a number of arithmetic operations proportional to $MN \log(MN)$. By using Perceval relation, we can obtain the correlation function by doing an FFT, evaluating the square modulus, and doing an Inverse FFT (IFFT). This only requires a number of operation of the order of $MN \log(MN)$. By scanning every value of Δx, and averaging over every $N \times M$ pixels, the number of operations would be of the order of $(MN)^2$. Using FFT, well known tricks can be used, in order to correct the boundary effects [19]. Figure 6.6 and 6.7 show the correlation functions thus evaluated.

The correlation function evaluated following the above described algorithm suffers from shot and read noise, that is, for the noise due to the CCD light measurement and acquisition systems. Since such a noise is not correlated to the speckle field due to scattered light, the noise correlation function sums to the speckle correlation function. In order to evaluate the noise correlation function, we acquire a set of about one hundred images, before putting the sample in the system. Then, we apply the above described algorithm to the images, and obtain the correlation function of the noise signal. Figure 6.8 shows the correlation function of the noise signal. We can notice a marked peak in 0, quite narrow, representing the correlation inside a row, and a correlation between lines spaced by two pixels, due to interlacing. The correlation function of the noise signal is

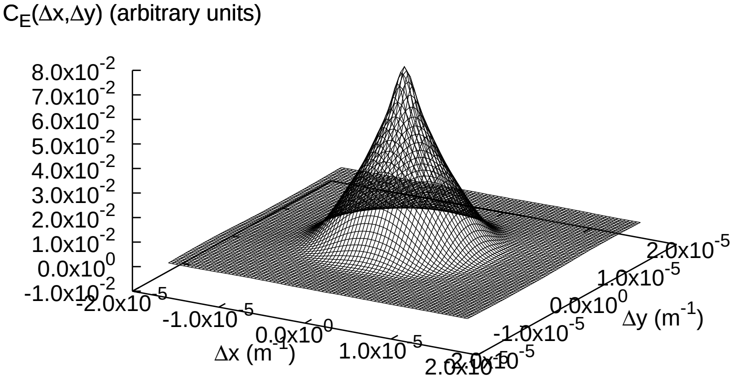

Figure 6.7: ENFS measurement of the field correlation function, for a 10.0μm colloid.

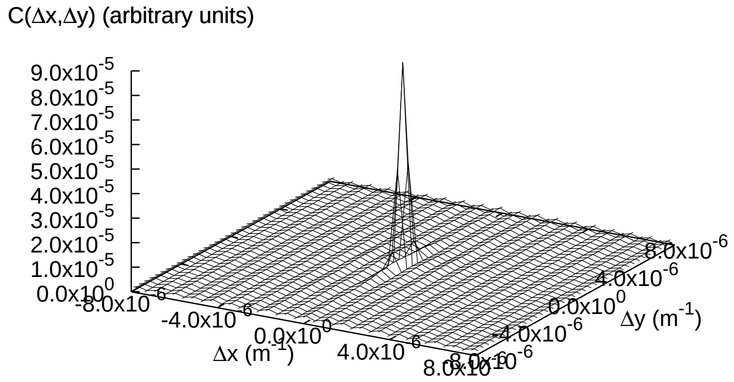

Figure 6.8: Correlation function of the shot and read noise.

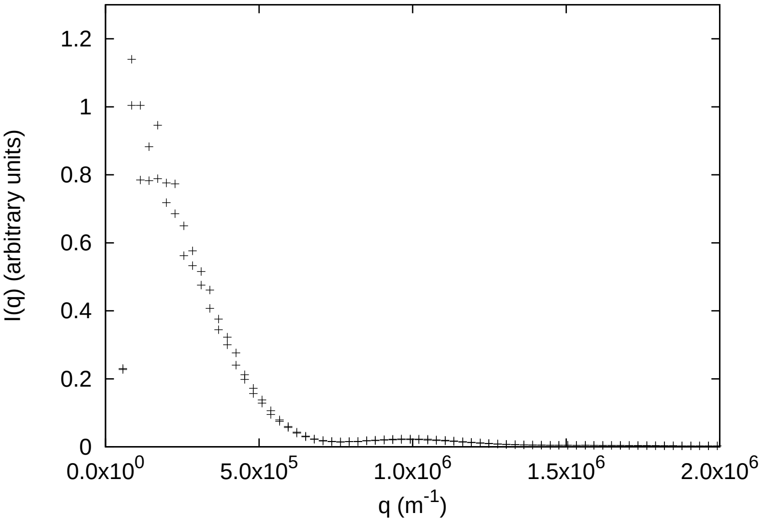

Figure 6.9: ENFS measurement of the scattered intensity of a 10.0μm colloid.

then subtracted by the overall correlation function.

Once the correlation function has been evaluated, through an FFT we obtain the field power spectrum $S_E(q)$. Since our samples are isotropic, we make an angular average of the power spectra, and represent our data as a function of the modulus q of $\vec{q}$. The scattered intensity $I(q)$ is then obtained by using Eq. (3.14), that is, simply relating each value of the power spectra, with wavelength q to a value of $I(Q)$, where the relation $Q(q)$ is given by Eq. (3.13). In Fig. 6.9 and 6.10 we show the measured $I(q)$.

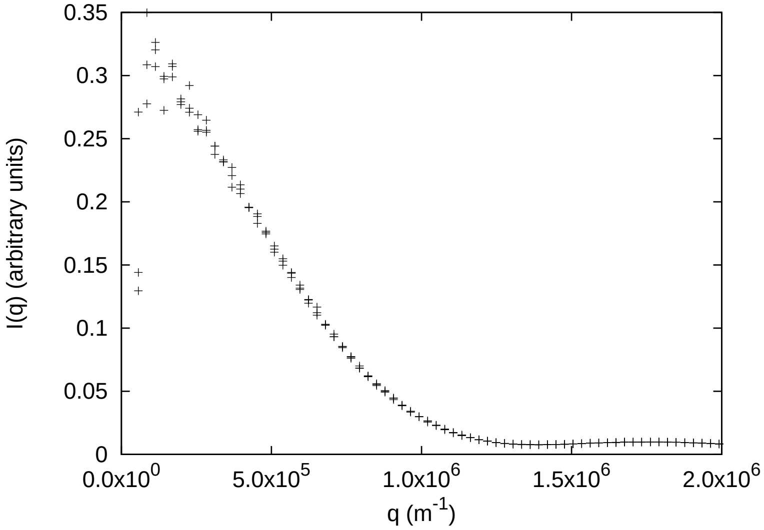

Figure 6.10: ENFS measurement of the scattered intensity of a 5.2μm colloid.