Fourth International Conference
Modelling and Development of Intelligent Systems
October 28 - November 1, 2015
"Lucian Blaga" University
Sibiu - Romania

# Using the Fourier Transform and the Power Spectral Density functions for Pattern Recognition in Dynamic Light Scattering Time Series

**Dan Chicea, Silviu Rei**

**Abstract**

If a coherent beam is incident on a suspension containing scattering centers, light is scattered and a far interference field is produced, having the aspect of speckled image. As the scattering centers undergo a complex motion, the far field presents fluctuations that carry information regarding the motion. The motion can be: a random Brownian motion, a relatively uniform sedimentation motion or a combination of both motions with different weight. Placing a detector in a far field and recording the fluctuations will produce a time series. A previously written and tested code named CHODIN was used to generate time series for several systems. The Fourier transform and the Power Spectral Density were used and the results are discussed in connection with the problem of very fast identification of the type of motion, and therefore the type of scattering centers suspended in the target fluid.

## 1 Introduction

The process known as Dynamic Light Scattering became more and more interesting in the last decades in the area of Physics and Chemistry. The physical process of light scattering on particles in a suspension can be used for implementing an efficient technique for determining the dynamics of the processes which take place in the suspension, with direct application in determining the size distribution of the particles. The main challenges in the first stages of the method were the difficulty to obtain a coherent monochromatic light beam and the computational complexity of the analysis required, rendering this technique unfeasible [1]. The emergence of LASERs and the increase of computing power in the last 20 years made this technique feasible.

Also known as "photon correlation spectroscopy" or "quasi elastic light scattering", this method involves a coherent monochromatic light source (in most of the situations a laser is used) which illuminates a sample containing a solution or suspension containing particles. Each particle undergoes an elastic interaction with the incident light beam and will act as secondary light source (Huygens principle) and will therefore scatter the light in all directions (Rayleight scattering for smaller particle sizes than the wavelength [2], [3], Mie scattering for bigger particles) [4]. The movement of the particles is, in the most general case, a complex phenomenon caused by the following concurrent processes: Brownian or thermal movement, aggregation and sedimentation.

Due to this complex movement each scattered light wave has a different phase leading to constructive, destructive or any other case in between by superposition, on a scattering image. Therefore the scattering image has spots of fluctuating intensity in each point and becomes a "boiling speckle" image. The intensity fluctuations contain information about the dynamics of the process, information which can be extracted by analysing the time series pattern recorded with a detector on a specific point. The analysis of the time series requires implementation of specific software which is able to recognise patterns in the input series offering as output information about several parameters of the suspension, like particle average size or size distribution.

In this paper we will focus on developing a very simple algorithm based on the Fourier transform and the Power Spectral Density, which is capable of analysing a time series obtained in a Dynamic Light Scattering experiment on particles having sizes in the range 250 – 1000 nm, due to the wide array of application of nano and micro particles in technology and sciences. The algorithm is able to indicate, at a qualitative level, which of the time series corresponds to smaller particles and which to bigger particles, or in other words, is capable to recognise patterns in the time series and to sort them based on particle size.

# 2 Fourier Transform and Power Spectral Density

The time series is recorded at discrete times, being therefore a stochastic or random discrete time series. Having in focus our target to design a simple algorithm capable of identifying patterns related to particle size, the periodicity of the stochastic time series was used for pattern recognition. For identifying the periodicities within the time series, one useful mathematical function is the Power Spectral Density which can be derived from the Fourier transform of the time series.

The Fourier transform is a reversible mathematical operation, which can transform a temporal signal from its time domain representation to its frequency domain representation. This operation is used for obtaining the frequency spectrum of a specific signal. For any real or complex function $f(t)$ which respects the condition:

$$\int_{-\infty}^{+\infty} f(t)dt < \infty \tag{1}$$

The truncated Fourier transform is defined as [5]:

$$F(\omega) = \frac{1}{\sqrt{2\pi T}} \int_0^T f(t)e^{-i\omega t}dt \tag{2}$$

For the specific case of this article, the time series was first averaged, than the average value was extracted and the new time series was squared, resulting the power time series of the signal. The Fourier transform defined above was used to produce the power spectral density, S(f), using the fast Fourier transform implemented in MATLAB.

# 3 Theoretical Heuristic Deduction

As described in the introduction, we will focus on several simulated time series produced during a DLS experiment by nano and microparticles. It has been shown [6] that the main process which influences the intensity fluctuations and therefore the variation of the time series recorded for nanoparticles is the Brownian motion. The bigger are the particles, the slower they move, fact that can be also seen in principle by inspecting equation below:

$$v = \sqrt{\frac{3k_B T}{m}} \tag{3}$$

Where $v$ is the root mean square speed, $k_B$ is Boltzmann`s constant, $T$ is the temperature and $m$ is the particle`s mass.

The fluctuation of the intensity is assumed to be more intense if the velocity of the particles is bigger, which means that the smaller particles are assumed to produce more intense fluctuations. Judging in frequency representation, this means that the higher frequencies have more weight than the lower frequencies in the Fourier transform for smaller particles time series (and the other way around for bigger particles). This means that the Power Spectral Density can be used as an indicator for identifying the pattern: particle size – frequency in the time series.

In this paper a very simple method for identifying this pattern is proposed. The power spectral density frequency domain is split into $n$ intervals:

$$0, f_1, f_2, \dots, f_n \tag{4}$$

An average for each interval is computed obtaining $n$ average values corresponding to the $n$ frequency ranges:

$$\langle S_1 \rangle, \dots, \langle S_n \rangle \tag{5}$$

The ratio of the lowest frequency to the highest frequency is calculated as:

$$R_i = \frac{\langle S_1 \rangle}{\langle S_n \rangle} \tag{6}$$

Data analysis presented in the next section reveals a monotone variation of the ratio R with the particle size, thus allowing the pattern recognition:

$$R_i \sim Size(i) \tag{7}$$

# 4 Algorithm Description

First a set of dynamic light scattering time series for different particle sizes was generated by using a previously developed and tested program called CHODIN [6]. One of the input parameters for CHODIN is the time step that is used to move the suspended particles at each simulation step. The algorithm for assessing the time step is described in detail in [7], therefore we will not repeat it here. First the time step for particles in the range 250-1000 nm was computed running the code described in [7]. For each particle size a set of input data was prepared for the CHODIN code described in detail in [7] and [8]. For each particle diameter, hence set of input parameters, a time series is generated by simulating Brownian motion in a volume and then calculating the intensity of scattered light at a specific angle and distance [7], [8]. Each time series is an input for the pattern recognition procedure.

For each time series the Power Spectral Density is calculated using the algorithm described in section 2.

In the next step the frequency domain of the Power Spectral Density is divided into $n$ intervals, each containing $m$ values. For each interval the average is calculated resulting in a series of $n$ averages, one for each interval, as described in section3. $R_i$ is calculated for each series using (6). Figure 1 illustrates the flow chart of the whole pattern recognition procedure.

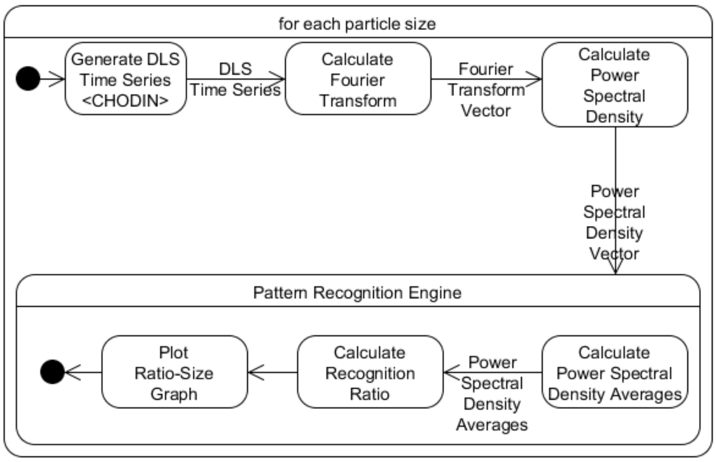


Figure 1 – the flow chart of the pattern recognition procedure

# 5 Results

Using the procedures described in section 4, DLS time series were generated for particles having a diameter of: 250, 375, 500, 625, 750, 825, 1000 (in nm). Each time series lasted for 1.5 s and were generated with 2000 data points per second. Figure 2 presents a sequence of 0.1 s of the time series generated for particles with a diameter of 250 nm and Figure 3 for particles with a diameter of 1000 nm.

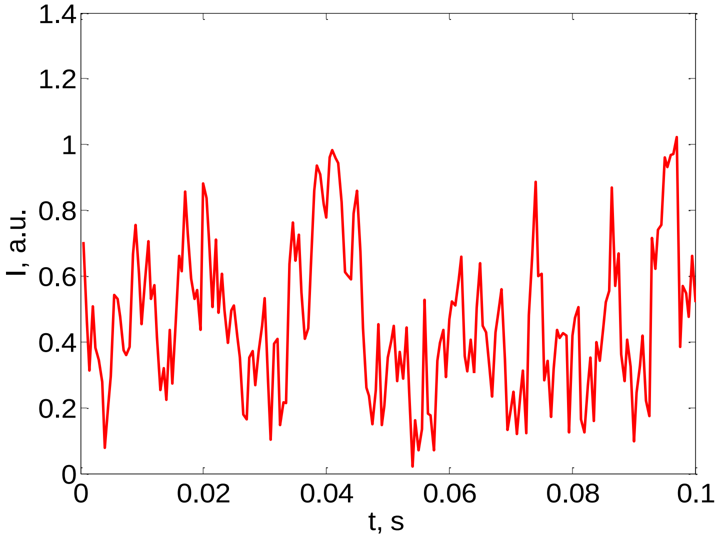


Figure 2 - a sequence of 0.1 s of the time series generated for particles with a diameter of 250 nm

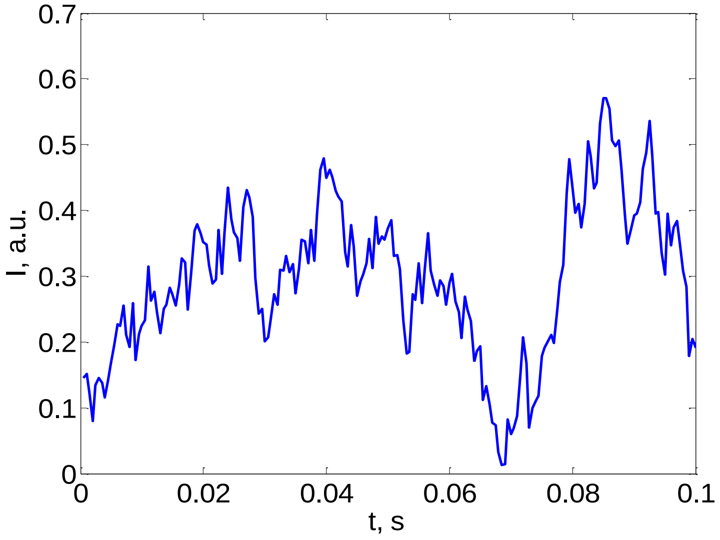


Figure 3 - a sequence of 0.1 s of the time series generated for particles with a diameter of 1000 nm

Examining Figures 2 and 3, we notice, even without any analysis tool, that the fluctuations in the time series produced by smaller particles have faster fluctuations with bigger amplitude. In order to step further from this quantitative observation, we compute the power spectral density as described in section 2. Moreover, we divided the total frequencies range in 10 intervals and computed the averages. Figure 4 illustrates the histogram of the averages of the power spectral density of the time series generated for particles having a diameter of 250 nm and Figure 5 for particles having a diameter of 1000 nm.

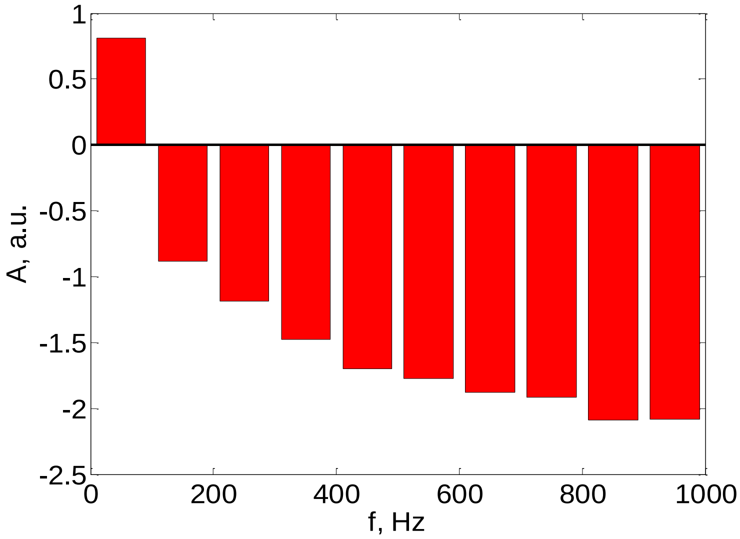


Figure 4 - The histogram of the power spectral density of the time series generated for particles having a diameter of 250 nm

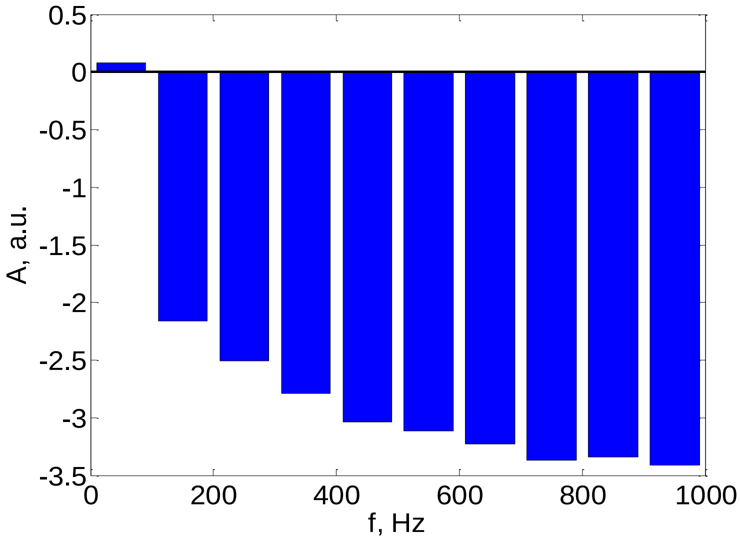


Figure 5 - The histogram of the power spectral density of the time series generated for particles having a diameter of 1000 nm

Again, at a qualitative level, we notice differences in the weight of different groups of frequencies in the power spectral density for the two power spectral densities.

The next step is to compute the ratio R using equation (6) for the time series mentioned at the beginning of this section. Figure 6 illustrates the variation of the ratio R with the diameter used to generate the DLS time series.

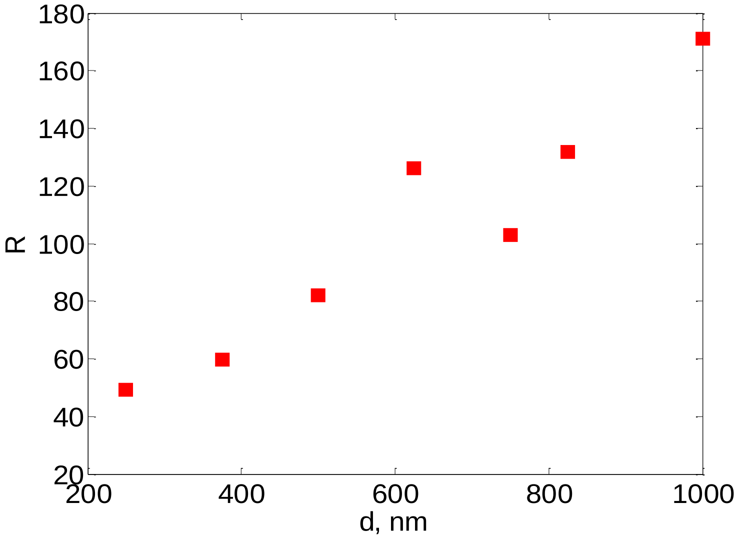


Figure 6 - The variation of the ratio R with the diameter used to generate the DLS time series

Examining Figure 6 we notice a monotone increase of the ratio R with the diameter, which confirms that smaller particles produce fluctuations in a DLS time series that have bigger frequencies.

# 6 Conclusion and discussions

DLS time series were generated in a realistic manner, using CHODIN, which moves each particle in suspension in an independent manner. The computer simulation of the Brownian motion of the particles in suspension is realistic, as the time step for the Brownian motion is chosen as a the value of a simulated diffusion experiment, that is the value that makes the simulated diffusion coefficient equal to the computed diffusion coefficient for that particular particle diameter and carrier fluid.

The seven time series present qualitative differences in respect of the fluctuations frequencies. The time series generated for smaller particles present faster fluctuations. A simple procedure using averages of the power spectral density on several frequency domains and computing the ratio R of the first (smallest frequencies) to the last (biggest frequencies) values of the averages was used. The results reveal a monotone increase of the ratio with the diameter of the suspended particles.

There are displacements though from the monotone variation, as can be noticed from Fig. 6 that presents the variation. These displacements might be the effect of the relatively small number of particles used in this simulation, 2000, and of the relatively small number of data in a set, 3000.

Nevertheless, the results are promising and suggest that a simple, easier and requiring a smaller amount of computer time, alternative procedure to DLS or Static Light Scattering [9] can be imagined, by calibrating a curve as suggested by the plot in Figure 6 and using it for assessing the diameter once the ratio R is computed for a recorded time series. The results presented here are partial results of the work that is in progress on this subject.

# References


[1] J.W. Goodman, *Laser speckle and related phenomena*, Vol.9 in series Topics in Applied Physics, J.C. Dainty, Ed., Springer-Verlag, Berlin, Heidelberg, New York, Tokyo, (1984).
[2] J. Strutt, *On the transmission of light through an atmosphere containing small particles in suspension, and on the origin of the blue of the sky*, Philosophical Magazine, 5(47), 375-394, (1899).
[3] A. T. Young, *Rayleigh scattering*, Appl. Opt. 20, 522–535, (1981).
[4] G. Mie. *Beiträge zur Optik trüber Medien, speziell kolloidaler Metallösungen,* Annalen der Physik **330** (3): 377–445, (1908).
[5] E. Hecht, Optics, 4-th Edition, Addison-Wesley, (2002)
[6] D. Chicea, *Coherent Light Scattering on Nanofluids – Computer Simulation Results*, Applied Optics, Vol. 47, No. 10, 1434-1442, (2008).
[7] D. Chicea, *A Simple Algorithm to Simulate Nanoparticles Motion in a Nanofluid*, , U.P.B. Sci. Bull., Series A, 76(2), 199-206, (`2014).
[8] D. Chicea, *Nanoparticle Sizing by Coherent Light Scattering – Computer Simulation Results*, Journal of Optoelectronics and Advanced Materials 10(4), 813-818, (2008).
[9] D. Chicea, *A Study of Nanoparticle Aggregation by Coherent Light Scattering*, Current Nanoscience, 8(6), 259-265, (2012), doi: http://dx.doi.org/10.2174/157341312800167704



Dan Chicea
University Lucian Blaga of Sibiu
Faculty of Sciences
Str. Dr. Ion Ratiu 7-9, Sibiu
Romania
E-mail: dan.chicea@ulbsibiu.ro

Silviu Rei
University Lucian Blaga of Sibiu
Herman Oberth Faculty of Engineering
Str. Emil Cioran 4, Sibiu
Romania
E-mail: silviu.rei@ulbsibiu.ro