

Counting and measuring micro and nano particles by using Fourier Interferometric Imaging

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ABSTRACT

This paper presents a new optical approach to measure micro and nano particles suspended in liquid with a very low concentration. The interference field created by light scattered from the particles on the surface of a 2D detector is recorded. The 2D FFT of this image maps the interacting pairs of particles. Therefore, the number, the 3D relative positions and sizes of the particles can be extracted.

Keywords: Lorenz-Mie theory, nano particle measurements, Brownian motion

1 INTRODUCTION

According with the European Community report [1], a Nano-material is defined as:

- *nanomaterial means a natural, incidental or manufactured material containing particles, in an unbound state or as an aggregate or as an agglomerate and where, for 50 % or more of the particles in the number size distribution, one or more external dimensions is in the size range 1 nm-100 nm.*
- *In specific cases and where warranted by concerns for the environments, health, safety or competitiveness the number of size distribution threshold of 50 % may be replaced by a threshold between 1 and 50 %.*

Consequently, the detection and measurement of nano particles with low concentration in a suspension containing largest particles is a challenge for fundamental as well as industrial issues.

To reach this aim, a new optical technique called Fourier Interferometric Imaging (FII) is proposed in this presentation. The main advantage of proposed approach is a higher sensitivity to small particles in the large control volume.

2 CONCEPTS

2.1 FII Concept

Fourier Interferometric Image (FII) approach has been introduced in [2]. Here we only recall the main basic

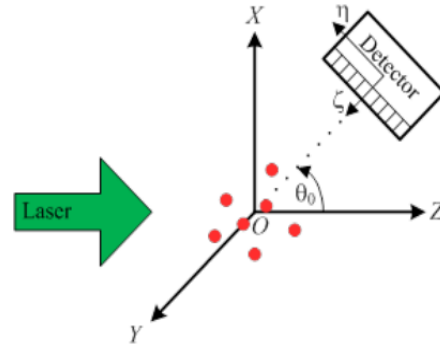


Figure 1: The problem under study

principle of this technique. Let us assume a cloud of particles illuminated by a pulsed laser and a CCD camera to collect the scattered light. On one pixel of the detector the total amplitude is given by:

$$A_t = \sum_{i=1}^N A_i \quad (1)$$

Where A_i is the amplitude of the light scattered by one particle and N is the total number of particles in the control volume. Accordingly, the total intensity on each pixel of the CCD camera is:

$$I_t = A_t \cdot A_t^* = \sum_{i=1}^N A_i A_i^* + \sum_{i=1}^N \sum_{j=1, j \neq i}^N A_i A_j^* \quad (2)$$

From equation [2], the first term is the total scattered intensity of each particle in the control volume. The second term is related to the interference between particles. FII is based on the exploitation of the second terms ($A_i A_j^*$).

To extract the terms ($A_i A_j^*$), a 2DFFT is used. The 2DFFT allows to separate the first term and second term. The information of the first term is located at the center of the 2DFFT image while all other traces on the 2DFFT image are the information of the second term. Figure 2 shows an example of the interference field of six silica particles and its associated 2DFFT. The following information can be extracted from the term of $A_i A_j^*$ identified as a trace in 2DFFT image:

- The distance between particles: the distance between a trace and the center of the 2DFFT image is a direct measurement of the distance between two particles.
- The number of particles: the number of traces is related directly to the number of particles in the control volume by $N(N - 1)$ where N is the number of particles in the control volume.
- Each particle is measured $N - 1$ times, where N is the number of particles in the control volume.
- The intensity of a trace is proportional to the square root of the product of the intensities scattered by each particle of interacting pair.

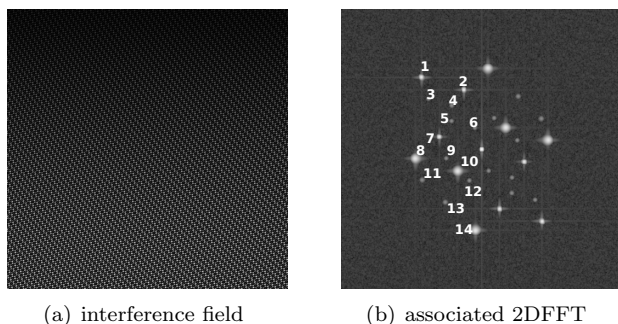


Figure 2: An interference field and its associated 2DFFT

3 NUMERICAL VALIDATION

3.1 Classical Brownian motion of a single particle

For an individual particle, the Brownian motion is characterized by the diffusion coefficient which is related to the mean squared displacement. Then the measurement of the mean squared displacement defined by:

$$\langle X_{(t)}^2 \rangle = \frac{\sum_{i=1}^M (x_{(t)} - x_{(0)})^2}{M} \quad (3)$$

where M is the number of measurements, $x_{(t)}$ is the particle location at time t and $x_{(0)}$ is the particle location at the first measurement. $\langle X_{(t)}^2 \rangle$ is a measurement of the particle size.

Starting from the text book of Akira Satoh, the Brownian motion has been simulated assuming a binomial distribution and a random number generation by using an adaptation of the Box-Muller algorithm.

Figure 3 presents a numerical example corresponding to a set of six silica particles (2 of diameter 20 nm and 4 of diameter 1 μm) in water with the integration and sample time steps equal to 1 ns and 0.01 sec respectively. The linear behaviour of $\langle X_{(t)}^2 \rangle$ versus time

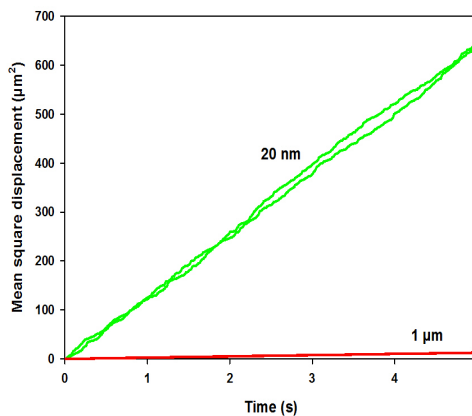


Figure 3: The mean squared displacement for individual particles of 20 nm (in green) and 1 μm (in red)

proves the quality of the Brownian movement simulations.

3.2 Distance between two Brownian particles

In FII approach, the particle location is not measured directly but the distance between two particles is measured. It is possible to show that the mean squared distance variation between two particles is related to the mean squared displacement of each particle by:

$$\langle X_{1,2}^2 \rangle = \langle X_1^2 \rangle + \langle X_2^2 \rangle \quad (4)$$

From the same series of numerical simulations used in figure 3, the squared distance variation has been computed from their coordinates. In this case of 6 particles, we must have 15 pairs of particles interacting two by two (see in figure 4). Then for each pair of particles the squared distance variation is computed and presented in figure 5. We observe that the squared distance variations of particles are gathered in 3 classes: the largest displacement corresponds to a pair 20 nm-20 nm, then the 8 pairs 20 nm-1 μm, and finally the 6 pairs 1 μm-1 μm.

This result proves that the variation of the distance between particles is a measurement of Brownian motion.

3.3 Brownian motion measurement by FII

FII works on 2DFFT of the interference field. The distance between particles is extracted from the location of the trace in the 2DFFT image. Then to validate the application of FII to Brownian particles, the interference field is computed by using the Lorenz-Mie code described in [3]. The same set of six silica particles is

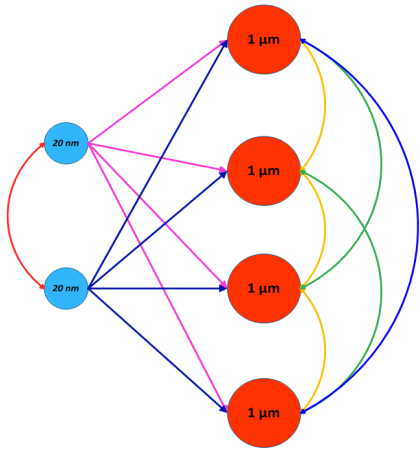


Figure 4: Interaction between six particles

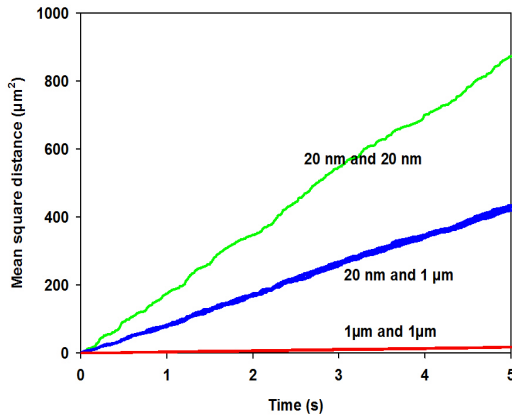


Figure 5: The mean squared distance variation between pairs of particle: pair of 20 nm particles (in green), pair of 20 nm and 1 μm (in blue) and pair of 1 μm (in red)

used to compute the interference field. Therefore, at each time step the interference field and its associated 2DFFT are computed as displayed in figure 2.

The variation of the distance between each pair of particles is extracted and then presented in figure 6. The linear regression is obtained which is similar to figure 5. This proves that FII technique is able to measure the mean squared distance variation between two particles, therefore, to characterize Brownian motion. However, we notice that the curve corresponding to a pair of particles of 20 nm-20 nm is missing. This fact is confirmed in figure 2(b) where only 14 traces are observed. The missing trace corresponds to the interaction between the particles of 20 nm and 20 nm. With this pair of particles, the interaction intensity is too low to be detected.

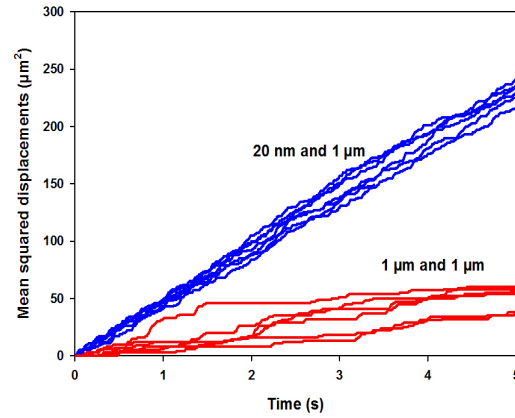


Figure 6: The mean squared distance variation measured from the 2DFFT of FII image. In blue 1 of 20 nm and one of 1 μm, in red two of 1 μm

3.4 FII signal intensity

The advantage of FII is to be able to measure small particles which cannot be seen by classical optical techniques. Often the optical techniques record the total scattered intensity of each particle but not the interference between particles. Then recorded scattered light is mainly dominated by big particles. Consequently, the measurement is weighted by the big particles.

The fact that FII focuses on the interference between particles permits to reduce the signal dependency on the particle size. Since trace intensity is the square root of the product of the scattered intensities from two interacting particles. Therefore, the detectability of small particles is increased. This is exemplified in Table 1 and figure 7.

Table 1 and figure 7 show the intensity of each trace on the 2DFFT image which corresponds to the interference between particles

Trace Number	η (pixel)	ζ (pixel)	Intensity
1	-107	129	18.210
2	-32	108	18.205
3	-96	91	11.268
4	-54	80	11.244
5	-54	49	11.274
6	-13	38	11.234
7	-78	22	18.192
8	-64	-17	11.244
9	-121	-17	17.830
10	-42	-40	17.830
11	-107	-55	11.182
12	-22	-59	11.269
13	-65	-97	11.172
14	-11	-147	17.874

Table 1: 2DFFT trace locations and intensities

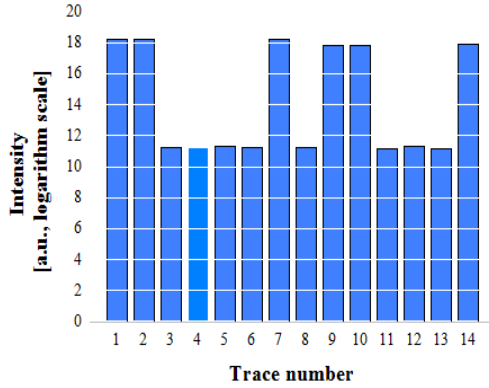


Figure 7: Trace intensity behavior

3.5 Particle individualization by triplet identification

To be able to measure each particle individually, the pair interaction must be identified. The identification process of three interacting traces (corresponding to three interacting particles) is exemplified in figure 8.

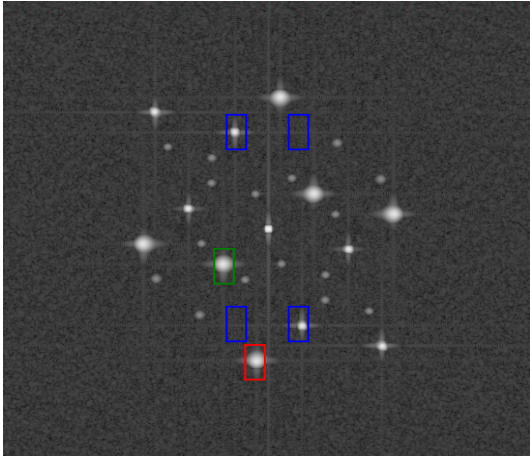


Figure 8: Triplet method to identify particles

A first trace is arbitrary selected, of coordinates η_1 and ζ_1 in the 2DFFT coordinates system (red rectangle in figure 8). Then a second trace is selected of coordinates η_2 and ζ_2 (green rectangle in figure 8). If a third interacting trace exists its coordinates are defined by [2]:

$$\eta_{1,2} = \pm(\eta_{1,3} - \eta_{2,3}) \quad (5)$$

$$\zeta_{1,2} = \pm(\zeta_{1,3} - \zeta_{2,3}) \quad (6)$$

The four rectangles satisfying these equations are plotted in figure 8 (in blue). If a trace is inside one of the four rectangles it means that this trace interacts with the two previously selected traces. Then the mean

squared distance variation corresponding to these three interacting traces can be determined given an equations system as follows;

$$\langle X_{1,2}^2 \rangle = \langle X_1^2 \rangle + \langle X_2^2 \rangle \quad (7)$$

$$\langle X_{1,3}^2 \rangle = \langle X_1^2 \rangle + \langle X_3^2 \rangle \quad (8)$$

$$\langle X_{2,3}^2 \rangle = \langle X_2^2 \rangle + \langle X_3^2 \rangle \quad (9)$$

where $\langle X_{1,2}^2 \rangle$, $\langle X_{1,3}^2 \rangle$, $\langle X_{2,3}^2 \rangle$ are measured.

The solution of these systems permits to measure the mean squared displacement of each individual particle. Finally, from classical Brownian motion equation the size of each individual particle can be obtained.

4 Conclusion

A strategy to apply the FII concept to the measurement of micro and nano particles has been proposed and numerically validated. The main advantage of the proposed approach is that the trace intensity does not depend on particle size in d^6 . This allows to distinguish the smallest particles in suspensions.

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