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Simulation of Photon Correlation Spectroscopy Signal Using Orthogonal Inverse Wavelet Transform

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Abstract

Computer simulation is a more convenient and faster method obtaining photon correlation spectroscopy (PCS) signal. Based on orthogonal inverse wavelet transform (OIWT), a new simulation method is developed. This method considers that PCS signal of a single scale is composed of several sub-band signals with different characteristic. According to the relationship of power spectrum of PCS signal and orthogonal wavelet coefficients of every scale, using OIWT, PCS signal can be obtained by simulation of several different sub-band signals. Using this method, PCS signals of 90nm, 600nm and1000nm are respectively simulated. Mean square errors of the power spectrums of the simulation signals and their theoretical power spectrums are e-5 order of magnitude. The relative errors of particle size inverted from simulation signals are less than 2.47%. Comparison of simulation and experiment proves that that OIWT is feasible for simulation of PCS signal. In addition, by analyzing the influence of simulation parameters on simulation accuracy, we get relationship of particle size, decomposition scale and sampling frequency.

Keywords: *power spectrum, photon correlation spectroscopy, orthogonal inverse wavelet transform, simulation signal*

1. Introduction

Photon Correlation Spectroscopy (PCS) is the most useful techniques for measuring size of nanoparticles and submicron particles in liquids, which often gets size information based on the measure of the temporal autocorrelation function (ACF) of scattered light intensity. Nowadays, this technique has applied in chemistry, physics and biological systems [1, 2]. In order to research characteristic of PCS signal, it is necessary to carry out extensive and accurate testing. Usually, the procedures are rigidly performed by using calibrated samples of latex spheres of different diameters. Although this procedure is well established, it presents several limitations and may lead to wrong or unclear results.

Compared with experiment method, PCS signal obtained by the computer simulation is more convenient, more flexible and faster. Although the generation of simulation signal of PCS has scarcely been investigated in the literature, the studies can be traced back to 1973. Hughes[3] firstly used the Gaussian–Markovian noises to simulate the scattered electric field, and then Shen and Ye [4-5] generated the PCS signal by the power spectrum. Later, based on the maximum likelihood principle, Lomakin [6] simulated the electric field scattered. Recently, WANG [7] also implemented the simulation of PCS signal by AR model. However, above all the simulation methods see PCS signal as a signal of single scale which is same characteristic. Therefore, simulation signals of these methods are coarser.

We consider that the PCS signal of a single scale is composed of several sub-band signals of multi-scale with different characteristics. Thus, PCS signal simulation of a single scale can be realized by simulation of several different sub-bands signal. Wavelet transform can decompose the signal into a few frequency bands, which is a new signal processing methods of good time-frequency characteristic. This method has been widely used for various engineering fields [8-10]. In addition, inverse wavelet transform also provides an effective method of reconstruction signal. In view of these causes, this paper proposes a simulation method of PCS signal based on orthogonal inverse wavelet transform (OIWT), which can simulate the light intensity fluctuations of nanoparticles and submicron particles.

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2. Characteristic Of PCS Signal

If the light field is a Gaussian distribution, its intensity is expressed as $I_s(t)$. $I_s(t)$ is a random process which fluctuates around the average value.The light intensity fluctuation is expressed as $< I_s(t) >$,which is a random process with zero means and relates to the particle size. The power spectrum of is expressed as [5]:

$$P(\omega) = \frac{2\Gamma / \pi}{\omega^2 + (2\Gamma)^2} \tag{1}$$

Where Γ relates to particle size and is called decay linewidth, ω is angular frequency.

The relations of decay linewidth and the particles size is expressed as [11]:

$$\Gamma = Dq^2, \quad q = \frac{4\pi n}{\lambda}\sin(\frac{\theta}{2}), \quad D = \frac{k_B \mathrm{T}}{3\pi\eta\, d} \tag{2}$$

Where d is the radius of the particles, k_B is the Boltzmann constant, T is the temperature in Kelvin degrees, η is the viscosity of the solvent, λ is the wavelength of the incident light in vacuum, n is the refractive index of the solvent and θ is scattering angle.

3. Simulation Principle of OIWT

3.1. Wavelet Transform Theory

Giving $\psi(t) \in L^2(R)$, $\psi(\omega)$ is Fourier transform of $\psi(t)$. If $\psi(\omega)$ satisfies $\int_{-\infty}^{\infty} \frac{|\psi(\omega)|^2}{|\omega|} d\omega < \infty$, then $\psi(t)$ is called the mother wavelet. The mother wavelet is stretched and shifted and can be expressed as:

$$\psi_{a,b}(t) = \frac{1}{\sqrt{|a|}}\psi(\frac{t-b}{a}) \tag{3}$$

Where $\psi_{a,b}(t)$ is called wavelet basis, a and b are the scaling parameters and shifting parameters, respectively.

Then the continuous wavelet transform of continuous function $x(t)$ is be expressed as:

$$W_f(a,b) = \frac{1}{\sqrt{|a|}}\int_{-\infty}^{\infty} x(t)\overline{\psi}(\frac{t-b}{a})dt \tag{4}$$

In practice, in order to reduce the redundancy of the wavelet transform coefficients, discrete wavelet transform of multi-resolution [12] is widely used. Its theory diagram is shown in Figure 1. Setting signal $x(t) \in V_0$, $x(t)$ can be decomposed into A_1 and D_1. A_1 corresponds to an approximation part of signal, which reflects low-frequency of signal. D_1 corresponds to detail part of signal, which reflects high-frequency of signal. Then, approximation part A_1 continues to be decomposed into A_2 and D_2. This process continues repeatedly until the decomposition scale reaches J scale, continuous signal $x(t)$ can decomposed into a few frequency bands and is expressed as:

$$x(t) = A_1 + D_1 = A_2 + D_2 + D_1 = A_3 + D_3 + D_2 + D_1 \tag{5}$$

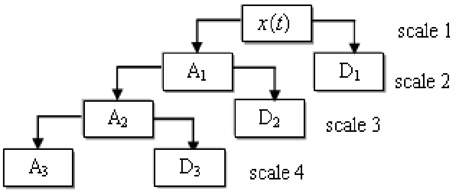

Figure 1. The Diagram of Wavelet Transform of Multi-resolution

Or $x(t)$ is written as:

$$x(t)=\sum_{j=1}^{J}\sum_{k=-\infty}^{\infty}d_{j,k}\psi_{j,k}(t)+\sum_{k=-\infty}^{\infty}c_{j,k}\phi_{j,k}(t) \tag{6}$$

Where $\phi_{j,k}(t)$ is scale function, c and d are the approximate coefficient and the detail coefficient, respectively. Formula (6) is called inverse wavelet transform. When the wavelet basis function $\psi_{j,k}(t)$ is the orthogonal basis and $J\rightarrow\infty$, $x(t)$ is only expressed by the details part as,

$$x(t)=\sum_{j,k}d_{j,k}\psi_{j,k}(t) \tag{7}$$

3.2. Relationship of Wavelet Coefficients and Power Spectrum

The average power of $x(t)$ can be expressed as:

$$\overline{P}=\lim_{T\rightarrow\infty}\int_{-\infty}^{\infty}\frac{1}{T}|x(t)|^{2}dt \tag{8}$$

Form Parseval theorem, we know:

$$\int_{-\infty}^{\infty}|x(t)|^{2}dt=\int_{-\infty}^{\infty}|X(f)|^{2}df \tag{9}$$

where $X(f)$ is Fourier transform of $x(t)$. According to formula (8) and (9) , formula (8) can be written as:

$$\overline{P}=\lim_{T\rightarrow\infty}\int_{-\infty}^{\infty}\frac{1}{T}|X(f)|^{2}df=\int_{-\infty}^{\infty}P(f)df \tag{10}$$

Where $P(f)=\lim_{T\rightarrow\infty}\frac{1}{T}|X(f)|^{2}$ is the power spectral density of $x(t)$.

According to multi-resolution ideas, the PCS signal of 2^{M} points can be approximately equal to approximation coefficient of the 1^{th} scale (most fine). When the sampling frequency is f_s, the number of coefficient and frequency range of the 2^{th} scale are 2^{M-1} and $[f_s/4, f_s/2]$,respectively. Accordingly, the number of coefficient and frequency range of the 3th scale are 2^{M-2} and $[f_s/8, f_s/4]$, respectively. So repeatedly, decomposition scale reaches J . Then, $x(t)$ is composed of the detail signals of all scales and approximation signal of most rough scale. For stationary random signal $x(t)$, the average is zero, when J is very large, the approximation signal of J^{th} scale tends to zero. Therefore, the signal $x(t)$ can be approximately expressed by the details signal of all scales. The signal $x(t)$ can be approximately expressed as:

$$x(t)=\sum_{j=1}^{J}\sum_{k=-\infty}^{\infty}d_{j,k}\Psi_{j,k}(t) \tag{11}$$

For orthogonal wavelet transform, on any j^{th} scale, the relationship of power spectrum and wavelet coefficients can be expressed as:

$$E_j=\sum_k d_{j,k}^2=2^M\int_{f_2}^{f_1}P(f)df \tag{12}$$

Where f_1, f_2 are low and upper limit of j^{th} scale frequency, respectively.

When the light field is Gaussian distribution, random fluctuations of light intensity $I(t)$ is the stationary random Gaussian process. For stationary random Gaussian process, on each scale, its wavelet coefficients is also the stationary random Gaussian process with zero means [13]. According to formula (12) and formula (1), we can be obtained the standard deviation of wavelet coefficients of every scale ($Var=\sum_k d_{j,k}^2 \Big/ 2^M$). Having standard deviation and mean of wavelet coefficients, wavelet coefficients of every scale can be easily generated by Gaussian white noises. According to wavelet coefficients of every scale, PCS signal satisfying target spectrum can be carried out by the inverse wavelet transform.

3.3. Simulation Algorithm

Steps of simulation algorithm are summarized as follows:

Step 1. Initializing particle radius d, sampling frequency f_s, decomposition scale J, simulation signal length $dataN$, experiment parameters T, η, λ, n, θ.

Step 2. According to the experiment parameters and formula (2), power spectrum $P(f)$ is calculated by formula (1) or formula (3).

Step 3. According to formula (13), the standard deviation Var of wavelet coefficients is calculated on every scale.

Step 4. According to Var of every scale, the wavelet coefficients of every scale are generated by Gaussian random noises.

Step 5. According to the wavelet coefficients of all scales, the signal is simulated by formula (12)

4. Signal Simulation and Analysis

In simulation experiments, simulation parameters are as follows: wavelength of incident beam is 632.8nm, refractive index of scattering medium is 1.331, scattering angle is 90°, temperature is 25℃, Botlzman constant is 1.3807×10^{-23} J·K^{-1}, viscosity coefficient of water is 0.89×10^{-3} N·S·m^{-2}, orthogonal wavelet basis is "db10".

4.1. Signal Simulation

At the same sampling frequency of 20kHz, the same simulation data length of 2^{19}, decomposition scale of 6, 12 and 17, respectively, the three kinds of PCS signals of 90nm, 600nm and 1000nm were performed by above the simulation method. The simulation signals with time range 0.1s are respectively shown in Figure 2. The power spectrum of every simulated signal were estimated by auto-regressive model [14]. Power spectrums of simulation signals and their theory power spectrums are shown in Figure 3. For every particle, mean square errors (MSE) of simulation power spectrums and their theory power spectrums are shown in Table1. According to Equation (1), three power spectrums of simulation signals were fitted by Levenberg-Marquardt algorithm [15]. Particles sizes were inverted from fitness results. The inverted results are also shown in Table1.

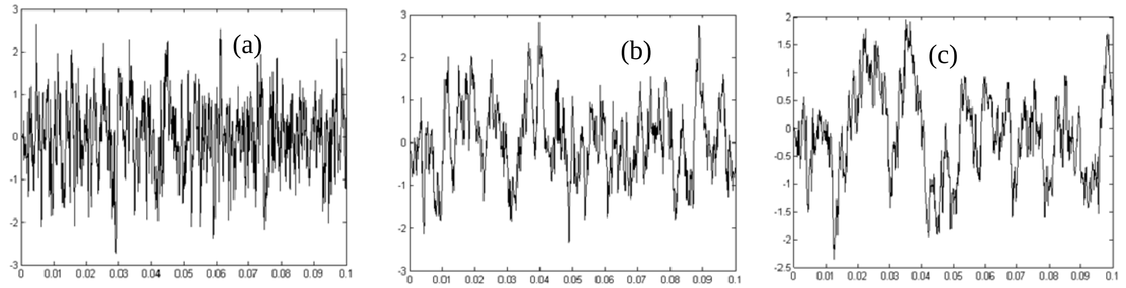

Figure 2. Simulation Signal of Particles (a) 90nm, (b) 600nm, (c) 1000nm

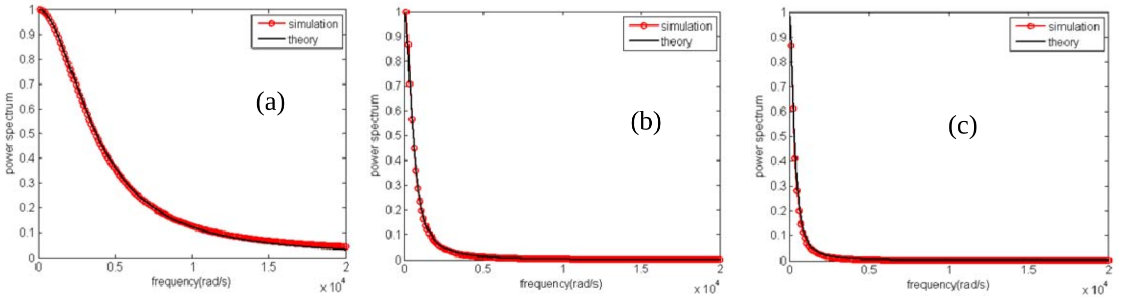

Figure 3. Power Spectrums of Simulation Signals and their Theory Power Spectrums

Table 1. Inverted Particles Size and MSE

Particle/nm	Inverted Size/Nm	Error%	MSE
90	91.17	1.30	5.1302e-5
600	590.76	1.54	1.9030e-5
1000	975.34	2.47	4.5120e-5

As can be seen from the Figure 2-3 and Table 1, the power spectrum of simulated signal performs a good agreement with its theoretical value. All MSE are e-5 order of magnitude. The size inversion relative errors of 90nm, 600nm and 1000nm are less than 2.47%. These results are shown that the simulation method based on OIWT is feasible for PCS signal.

4.2. Parameters Influence Analysis

4.2.1. Influence of Decomposition Scale

At two kinds of simulation parameters with f_s =10kHz, $dataN$ =2^{18}, J =6; f_s =10kHz, $dataN$ =2^{18}, J =17, PCS signal with particles sizes 100nm, 300nm and1000nm were simulated respectively. The power spectrums of simulation signals and their theory power spectrums are shown in Figure 4-5. For every case, MSE of two power spectrums and inverted particle sizes from the power spectrums of simulation signals are shown in Table 2.

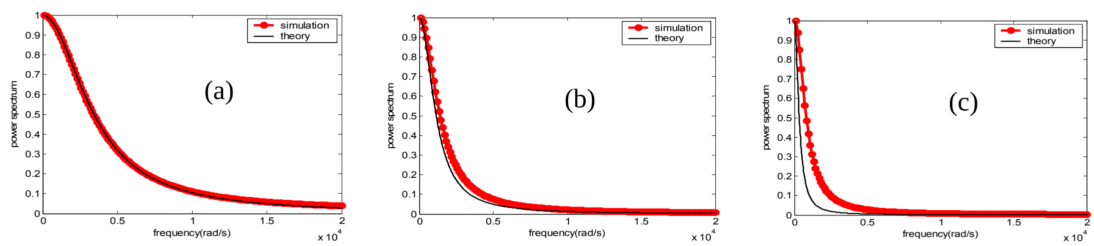

Figure 4. Power Spectrums of Simulation Signals and their Theory Power Spectrums with Parameters f_s =10kHz, $dataN$ =2^{18}, J =6 (a) 100nm (b)300nm(c)1000nm

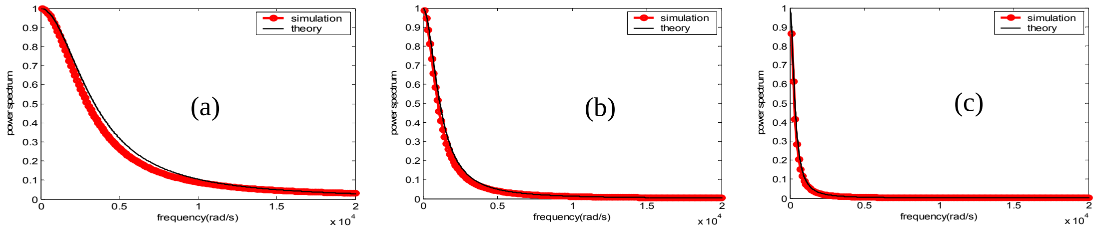

Figure 5. Power Spectrums of Simulation Signals and their Theory Power Spectrums with Parameters f_s =10kHz, $dataN$ =2^{18}, J =17 (a) 100nm, (b) 300nm, (c)1000nm

Table 2. Inverted Particles Size and MSE

Particle/nm	J=6			J=17		
	Inverted Size/Nm	Error%	MSE	Inverted Size/Nm	Error%	MSE
100	101.92	1.92	6.1321e-5	107.15	7.15	8.0423e-4
300	280.17	6.61	7.5679e-4	308.46	2.82	9.2822e-5
1000	875.23	12.48	4.6125 e-3	976.29	2.37	4.3017e-5

From Figures 4-5 and Table 2, we can find out that, at same sampling frequency and the data length, when decomposition scale increases, the power spectrums of three simulated signals doesn't always agree with its theoretical value very well, at smaller decomposition scale, the MSE and errors of the small particles of 100nm are smaller, corresponding simulation signal has higher simulation accuracy. On the contrary, for the big particles of 300nm and 1000nm, simulation accuracy is higher at the bigger decomposition scale. Reason of this phenomenon is as follow: particle size information mainly contains in the decay part of power spectrum. For big particles, decay part of power spectrum is in the narrower low frequency band, when decomposition scale is too small, according to simulation principle of OIWT, theory power spectrum of simulation signal is divided into less number of frequency bands, at same sampling frequency, width of every frequency band is bigger, thus decay part of power spectrum relating to particle size contains few sampling points, particle size information is lost a lot and leads to poor simulation accuracy. For small particles, decay part of power spectrum is wider frequency band, when decomposition scale is very large, power spectrum contains enough sampling points, however, distribution of these sampling points is very uneven, a large number of sampling points are near zero frequency of power spectrum, decay part relating to particle size has fewer sampling points, accordingly leads to poor simulation accuracy. Therefore, in simulation of PCS signal, we should choose the bigger decomposition scale for big particles and the smaller decomposition scale for small particles.

4.2.2. Influence of Sample Frequency

PCS signals of 100nm were simulated at same data length of 2^{18}, same decomposition scale of 6, different sampling frequencies f_s =10kHz, f_s =20kHz, f_s =5kHz, respectively. The power spectrums of simulation signals and their theory power spectrums are shown in Figure 6-7. MSE of two power spectrums and inverted particle size from the power spectrum of simulation signal are shown in Table 3.

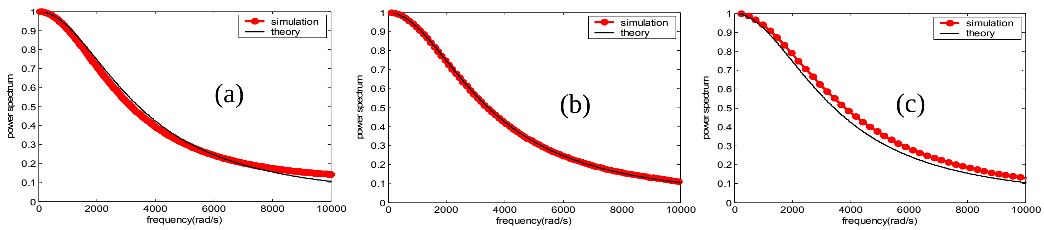

Figure 6. Power Spectrums of Simulation Signals and their Theory Power Spectrums of 100nm at Different Sample Frequencys (a) f_s =5kHz (b) f_s =10kHz, (c) f_s =20kHz

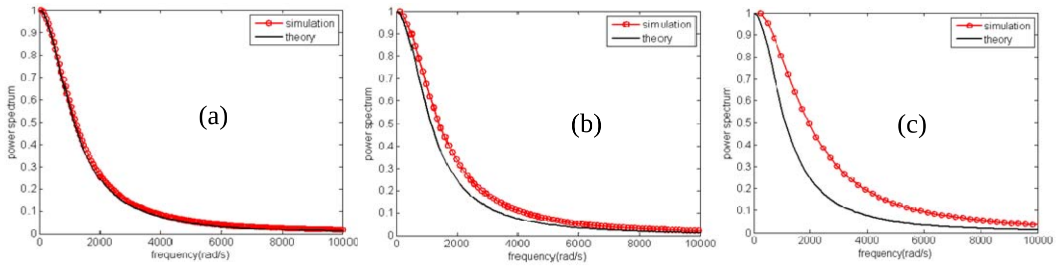

Figure 7. Power Spectrums of Simulation Signals and their Theory Power Spectrums of 300nm at Different Sample Frequencys (a) f_s =5kHz (b) f_s =10kHz (c) f_s =20kHz

Table 3. Inverted Particles Size and MSE

Particle/nm	100nm			300nm		
	inverted size/nm	error%	MSE	inverted size/nm	error%	MSE
5Hz	108.41	8.41	6.0294e-004	296.47	1.17	1.1036e-005
10kHz	98.12	1.88	8.0874e-005	283.16	5.61	7.4087e-004
20kHz	115.87	15.87	2.6720e-004	256.36	14.54	0.0026

As can be seen from Figure 6-7 and Table 2, at same data length and decomposition scale, when sampling frequency is too large or too small, MSE of two power spectrums and corresponding inversion size errors are bigger. Only at appropriate sampling frequency of 10kHz can simulation signals have higher accuracy. When sampling frequency is too small, decay part of power spectrum can't be completely sampled and lost a lot of particle size information, simulation accuracy is poor. When sampling frequency is too large, frequency band of every scale is very wide at same decomposition scale, decay part of power spectrum contains few sampling points and simulation accuracy is also poor. Therefore, the appropriate sampling frequency should be chosen in the simulation. Generally speaking, different particle have different appropriate sampling frequency, appropriate sampling frequency is related to particle size. For big particles, decay part of power spectrum is narrower, appropriate sampling frequency is smaller, for small particles, it is contrary.

In practice, the two parameters are mutually constraints. We should synthesize influence of two parameters to choose the appropriate sampling frequency and decomposition scale

5. Simulations and Experiment

In order to test effectiveness of simulation signal obtained by OIWT method, we compare the experimental statistics of field scattered by particles system with that resulting from our simulation method. The experiment setup implemented by our research groups [16] is shown in Figure 8, which mainly involves a He-Ne laser with a wavelength of 632.8nm, FPGA chip of digital signal processing. Experimental photon signal and correlation function can get by FPGA chip. The experiment materials were standard polystyrene latex spheres which were suspended in purified water. The measurements were implemented at sample time of 50μs, scattering angle of 90° and temperature of 298$\pm$2. The simulation signals of particles size 200nm were obtained by OIWT method. In simulation experiment, experiment parameters are the same as the section 4 and simulation parameter are f_s =20kHz, $dataN$ =2^{18} and J =12, respectively. After obtaining simulation and measurement signal, we calculated power spectrum of two signals, and compared them. Two power spectrums are shown Figure 9. From Figure 9, an excellent accord between experiment power spectrum and simulation power spectrum is found. The decay part and the temporal ranges of two power spectrums are obvious agree completely. Furthermore, two power spectrums were inverted. The inverted average sizes of simulation power spectrum and measurement power spectrum are 203.95nm and 204.52nm.

Two inverted results also agree. In summary, these results are certainly encouraging. It indicates that OIWT method can be used for PCS signal simulation of nanoparticles and submicron particles system.

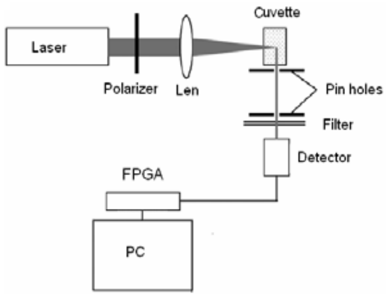

Figure 8. Experimental Setup

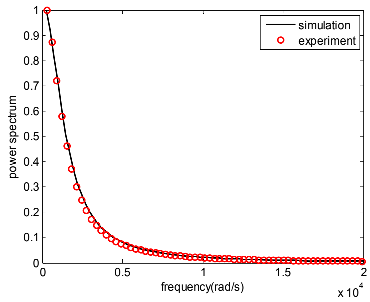

Figure 9. Power Spectrum of Simulation and Experiment Signal

6. Conclusion

According to the relationship of power spectrum of fluctuating light intensity and wavelet coefficients of every scale, based on OIWT, this paper proposes a simulation method of PCS signal.This method considers characteristics of different sub-band signals, and is multi-scale method which overcomes the shortcomings of coarse simulation in the general single scale method. By this method, the PCS signals of particles with particles sizes 90nm, 600nm and 1000nm were respectively simulated. The power spectrums of simulative signals perform great agreement with their theoretical values. MSE of two power spectrums is e-5 order of magnitude. The inversion size errors of simulation signals are less than 2.47%. Therefore, OIWT method is feasible for simualtion of PCS signal. In addition, this paper studies the influence of simulation parameters of decomposition scale and sample frequency on simulation accuracy. Research shows that the small particles need smaller decomposition scale and sampling frequency, big particles have contrary conclusion. Comparison of simulations and experiment prove that OIWT method can be used for PCS signal simulation.

Acknowledgement

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