Measurements of Particle Size Distribution Based on Mie Scattering Theory and Markov Chain Inversion Algorithm

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***Abstract*—Measuring particle size distribution through calculating light scattering intensity is a typical inverse problem. This paper builds an inverse mathematical model based on Mie scattering, deduces the inversion formulas for particle size, and calculates the relative coefficients through programming with built-in functions in MATLAB. In order to improve the accuracy and noise immunity of particle size distribution measurement, the development of stochastic inversion algorithm: an inverse problem model based on Markov chain algorithm is proposed. Results of numerical simulation are added acceptable noise indicate that the algorithm of Markov chain has strong noise immunity and can meet the requirements of on-line measurement.**

***Index Terms*—particle size distribution, Mie scattering theory, inversion algorithm, Markov chain, light scattering method**

I. Introduction

In recent years, the research of particle size measurement techniques focuses on the method based on light scattering theory. Light scattering is the most widely method used in particle size measurement, which is characterized by the fast testing speed, high precision, good repeatability, high resolution, wide applicability and wide range of particle size measurement. The little unknown physical parameters of measured particle and dispersion medium, and high degree of automation and intelligence of instruments make on-line measurement could be realized. Light scattering method is based on light scattering theory: The light beam will scatter around the space when incidents on the measured particles, the scattering parameters are closely related to particle size distribution. The scattering parameters which can affect the particle size are: spatial distribution of scattering intensity, the attenuation coefficient of transmission light intensity relative to incident light, degree of polarization of scattered light and so on. The ideal particle size distribution can be obtained by measuring of relevant scattering parameters and their combination. The workflows applied to the particle measuring instrument are basically the same. When the light beam irradiates testing particles in a space, there would produce the light scattering which can be converted into electrical signals through optical detector, these signals could be amplified by the amplifier and then processed by the computer, and after these steps the information about particle size distribution can be obtained. In this article the particle size distribution is assumed to obey a particular function, and then obtained by inversion of measuring data. Particle size and particle concentration are the two main issues in the research of particle measurement. In fact, one of them can be calculated from the other. In this paper Markov chain algorithm is applied to the inverse problem of particle size measurement based on light scattering theory. By comparing fitting extent of particle size distribution results with given distribution function under noise and no-noise, the research of algorithm noise immunity could apply a basis for the online measurement.

II. Comparison with Existing Research

In various measuring methods, particle is discussed based on the assumption of which is spherical particle. Most standard particles are spherical and there has a high requirement for spherical degree. The standard particle is usually the monodisperse particle, which has very narrow particle size distribution. The light scattering method, a non-disturbance detection technology, used to measure particle size distribution has shown its good prospects with its unique superiority, as the particle and powder materials widely applied in national defense, medicine, technology and manufacturing, accompanied by the developing of laser, optical and computer technology.

Particle size analysis is based on the inversion of a diffusion matrix, the resolution of which is based on theories stemming from the equations of Maxwell. At present, the main mature theories of light scattering are: Mie scattering theory, Fraunhofer diffraction theory, and Rayleigh scattering theory. There is a wide research on the measurement technique of particle size distribution

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based on Mie scattering theory and Fraunhofer diffraction theory. The Mie theory and the Fraunhofer diffraction theory approximation enable particle size distributions between several tens of nanometers and several thousands of micrometers to be calculated. The Fraunhofer diffraction equation is a simplified version of the Kirchhoff's diffraction formula and it can be used to model the light diffracted when both the light source and the viewing plane are effectively at infinity with respect to the diffracting aperture. Fraunhofer can be approximately seen as the simplified model of Mie theory, so it has lower precision than Mie theory. Rayleigh scattering, named after the British physicist Lord Rayleigh, is the elastic scattering of light or other electromagnetic radiation by particles much smaller than the wavelength of the light. The particles may be individual atoms or molecules. It can occur when light travels through transparent solids and liquids, but is most prominently seen in gases[1][2].

Comparison of results, Mie theory is more suitable for measurements of particle size. To measure particle size using algorithm based on Mie theory can be applied to various types of particles and has a higher accuracy than other algorithm

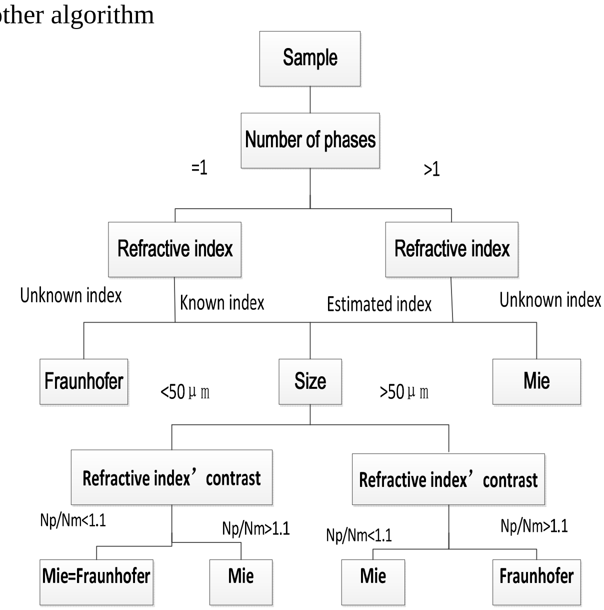

Figure 1. The comparison between Mie scattering and Fraunhofer diffraction

The Fig. 1 shows, in the case of small or transparent particles, that the use of the Fraunhofer approximation leads to an error in the size measurement. The main consequences and limits of Fraunhofer used in the model are the presence of populations which are actually absent or a shift in the particle size distribution towards larger particles.

In this article the particle size distribution is assumed to obey a particular function, and then obtained by inversion of measuring data. Particle size and particle concentration are the two main issues in the research of particle measurement. In fact, one of them can be calculated from the other. In this paper Markov chain algorithm is applied to the inversion problem of particle size measurement based on Mie scattering theory. By comparing fitting extent of particle size distribution results with given distribution function under noise and no-noise, the research of algorithm noise immunity could apply a basis for the online measurement. In various measuring methods, particle is discussed based on the assumption of which is spherical particle. Most standard particles are spherical and there has a high requirement for spherical degree. The standard particle is usually the monodisperse particle, which has very narrow particle size distribution. The light scattering method, a non-disturbance detection technology, used to measure particle size distribution has shown its good prospects with its unique superiority, as the particle and powder materials widely applied in national defense, medicine, technology and manufacturing, accompanied by the developing of laser, optical and computer technology.

III. Measuring Model Based on Mie Scattering Theory

A. *Representation of Inversion Problem*

The light scattering phenomena will occur when a beam of monochromatic parallel light, which has light intensity I_0 and wavelength λ, irradiates a spherical particle with radius a, and scattering intensity is $K(\theta,\lambda,a)$ in space angle θ. Particles are idealized to be strictly spherical in the measuring research of particle group size. To assume the frequency distribution function of particle size is $f(a)$ without any other effect, the total number of particles is N, then

$$I(v)=\int_{a_{\min}}^{a_{\max}} K(v,a)f(a)da \quad . \qquad (1)$$

Here v is θ or λ, corresponding to angle spectrum method and light spectrum method respectively. Angle spectrum method is to measure the distribution of light scattering intensity in space under a certain wavelength, while the light spectrum method is under a certain angle[3]. This paper focuses on the method of angle spectrum. Light intensity data $I(v)$ can be measured by measuring device, and the inverse problem of particle size measurement is to obtain the expression of particle size distribution $f(a)$ from $I(v)$.

B. *Solving for Parameters*

First to discuss the case of that the incident light is completely polarized light, assuming the incident light irradiates along the z-axis, the electric vector is along the x-axis, I_r, I_l, the scattering intensity which is perpendicular, parallel to the scattering plane and the total intensity I_s are

$$I_r=\frac{\lambda^2 I_0}{4\pi^2 r^2}\left|s_1(\theta)\right|^2 \sin^2\varphi=\frac{\lambda^2 I_0}{4\pi^2 r^2} i_1(\theta)\sin^2\varphi \qquad (2)$$

$$I_l = \frac{\lambda^2 I_0}{4\pi^2 r^2}|s_2(\theta)|^2 \cos^2\varphi = \frac{\lambda^2 I_0}{4\pi^2 r^2} i_2(\theta)\cos^2\varphi \quad (3)$$

$$I_s = \frac{\lambda^2 I_0}{4\pi^2 r^2}[i_1(\theta)\sin^2\varphi + i_2(\theta)\cos^2\varphi]. \quad (4)$$

The schematic diagram of Mie scattering as follows:

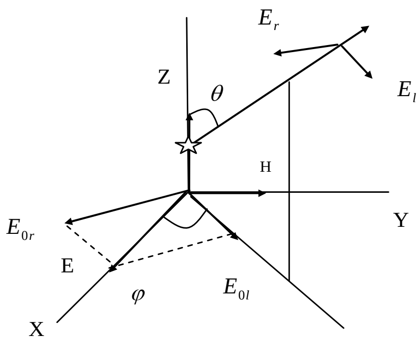

Figure 2. Mie scattering schematic diagram

θ is scattering angle, r is the distance between scattered particles and measuring point P, $i_1(\theta)$ and $i_2(\theta)$ are called scattering intensity function. φ is the angle between vibrating surface of incident light and scattering surface.

According to Mie scattering theory, the scattering amplitude function is the infinite series composed by Bessel function and Legendre function[3][4], which can be expressed as

$$S_1(\theta) = \sum_{L=1}^{\infty}\frac{2L+1}{L(L+1)}(a_L\pi_L + b_L\tau_L) \quad (5)$$

$$S_2(\theta) = \sum_{L=1}^{\infty}\frac{2L+1}{L(L+1)}(a_L\tau_L + b_L\pi_L) \quad (6)$$

a_L, b_L are all related to particle size, and π_L, τ_L are all related to scattering θ.

$$a_L = \frac{\varphi_L(x)\varphi_L'(mx) - m\varphi_L'(x)\varphi_L(mx)}{\xi_L(x)\varphi_L'(mx) - m\xi_L'(x)\varphi_L(mx)} \quad (7)$$

$$b_L = \frac{m\varphi_L(x)\varphi_L'(mx) - \varphi_L'(x)\varphi_L(mx)}{m\xi_L(x)\varphi_L'(mx) - \xi_L'(x)\varphi_L(mx)} \quad (8)$$

$$\pi_L = \frac{P_L^{(1)}(\cos\theta)}{\sin\theta} \quad (9)$$

$$\tau_L = \frac{d}{d\theta}P_L^{(1)}(\cos\theta) \quad (10)$$

The parameter of particle size $\alpha = 2\pi a/\lambda$, $P_L^{(1)}$ is first-order and L-time Lagendre fuction. $\varphi_L(z)$ and $\xi_L(z)$ are Ricatti-Bessel function, they are half-integral order Bessel function and Hankel function of the second kind (z could be α or $m\alpha$)[1][2][3][5]. $\varphi'_L(x)$ and $\xi'_L(x)$ denote the differential quotient of $\varphi_L(x)$ and $\xi_L(x)$ for each independent variable respectively.

$$\xi_L(z) = \varphi_L(z) + i * \chi_L(z) \quad (11)$$

$$\varphi_L(z) = \sqrt{\frac{\pi z}{2}} J_{L+1/2}(z) \quad (12)$$

$$\chi_L(z) = -\sqrt{\frac{\pi z}{2}} H^{(2)}{}_{L+1/2}(z) \quad (13)$$

To assume the scattering amplitude function $S_1(\theta)$, $S_2(\theta)$ and the scattering intensity function $i_1(\theta)$, $i_2(\theta)$ have the relationship as

$$i_1(\theta) = |S_1(\theta)|^2 \quad (14)$$

$$i_2(\theta) = |S_2(\theta)|^2 \quad (15)$$

Solving the scattering coefficients a_L, b_L and π_L, τ_L is the key of seeking for the scattering intensity function $i_1(\theta)$ and $i_2(\theta)$.

If the incident light is natural light, the scattered light is partially polarized light, so the scattering intensity $I(\theta)$ and polarization P are

$$I(\theta) = I_0 \frac{\lambda^2}{8\pi^2 r^2}(i_1(\theta) + i_2(\theta)) \quad (16)$$

$$P = \frac{i_1(\theta) - i_2(\theta)}{i_1(\theta) + i_2(\theta)}. \quad (17)$$

Whether the incident light is polarized or natural light, the efficiency factor K_{scat}, extinction efficiency factor K_{ext} and absorption efficiency factor K_{abs} of particle scattering are

$$K_{sca} = \frac{2}{\alpha^2}\sum_{L=1}^{\infty}(2L+1)\left[|a_L|^2 + |b_L|^2\right] \quad (18)$$

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$$K_{ext} = \frac{2}{\alpha^2}\sum_{L=1}^{\infty}(2L+1)\,\mathrm{Re}(a_L + b_L) \quad (19)$$

$$K_{abs} = K_{ext} - K_{sca}\,. \quad (20)$$

The calculation of Mie coefficients a_L and b_L is the initial problem of the calculating for the physical quantity related to light scattering. The scattering coefficient, extinction coefficient and absorption coefficient can be obtained by formula of Mie coefficients. The calculation of scattering amplitude functions $s_1(\theta)$ and $s_2(\theta)$ also depends on Mie coefficients calculating. Scattering intensity function $i_1(\theta)$ and $i_2(\theta)$ can be obtained through numerical calculation by a known particle size α and refractive index m of particle materials. Wavelength of incident light affects scattering light through changing dimensionless parameter α and relative refractive index m. For the particles with small conductivity σ, m has very little to do with incident light frequency or wavelength, and the wavelength works through α, essentially the effect caused by wavelength changing is equal to appropriate changing of particle size. For the particles with big conductivity σ, value of m differs widely under different incident light frequency, and dimensionless parameter α and relative refractive index m are all affected by wavelength changing.

C. Calculation of Relative Coefficients in MATLAB

Using built-in commands and setting functions in MATLAB (Half-odd-order Bessel function of the first kind $J_{L+1/2}(x)$ and the second kind $H_{L+1/2}(x)$ can obtain accurate results of spherical particles scattering parameters in any refractive index and a wide scale range[5][6].

To make some simple deformations for (7) and (8), dividing the right numerator and denominator by $\varphi_L(m\alpha)$.

$$a_L = \frac{\varphi_L(\alpha)\dfrac{\varphi_L'(m\alpha)}{\varphi_L(m\alpha)} - m\varphi_L'(\alpha)}{\xi_L(\alpha)\dfrac{\varphi_L'(m\alpha)}{\varphi_L(m\alpha)} - m\xi_L'(\alpha)} \quad (21)$$

$$b_L = \frac{m\varphi_L(\alpha)\dfrac{\varphi_L'(m\alpha)}{\varphi_L(m\alpha)} - \varphi_L'(\alpha)}{m\xi_L(\alpha)\dfrac{\varphi_L'(m\alpha)}{\varphi_L(m\alpha)} - \xi_L'(\alpha)} \quad (22)$$

The recurrent relationship between $\varphi_L(x)$, $\chi_L(x)$ and their differential quotient is given as

$$\left.\begin{aligned}
\varphi_L(x) &= \frac{2L-1}{x}\varphi_{L-1}(x) - \varphi_{L-2}(x)\\
\varphi'_L(x) &= -\frac{L}{x}\varphi_L(x) + \varphi_{L-1}(x)\\
\chi_L(x) &= \frac{2L-1}{x}\chi_{L-1}(x) - \chi_{L-2}(x)\\
\chi'_L(x) &= -\frac{L}{x}\chi_L(x) + \chi_{L-1}(x)
\end{aligned}\right\} \quad (23)$$

Thus

$$\begin{cases}
\varphi'_L(x) = \varphi_{L-1}(x) - \dfrac{L}{x}\varphi_L(x)\\
\chi'_L(x) = \chi_{L-1}(x) - \dfrac{L}{x}\chi_L(x)
\end{cases} \quad (24)$$

The assumed initial conditions in numerical simulation are $\varphi_0(z) = \sin z$, $\varphi_{-1}(z) = \cos z$, $\chi_0(z) = \cos z$, $\chi_{-1}(z) = -\sin z$, $\tau_1 = \cos\theta$ and $\pi_1 = 1$, substituted into relative formulas and a_L, b_L can be deduced, the scattering angle function π_L, τ_L are related to scattering angle θ, so the recurrent relationship is

$$\pi_L = \frac{2L-1}{L-1}\cos\theta\pi_{L-1} - \frac{L}{L-1}\pi_{L-2} \quad (25)$$

$$\pi'_L = \frac{2L-1}{L-1}\pi_{L-1} + \frac{2L-1}{L-1}\cos\theta\pi'_{L-1} - \frac{L}{L-1}\pi'_{L-2} \quad (26)$$

Through the setting of symbol variable x and symbol function $\varphi_L(x)$, $\xi_L(x)$, then making derivation and deformation for each side of (11), it can be deduced as follow

$$\xi_L'(x) = \varphi_L'(x) + i*\chi_L'(x) = \xi_{L-1}(x) - \frac{L}{x}\xi_L(x) \quad (27)$$

To transform symbol variable and symbol function into numeric variable α and $m\alpha$ numeric function using the buit-in function Inline and Vectorize in MATLAB, a_L, b_L can be deduced through substituting the variable value, and then calculate π_L, τ_L using built-in function Legend.

In order to demonstrate the feasibility of this method, to plot the relationship between extinction coefficient and particle size under different refractive index, and the accuracy of a_L, b_L can be verified through this method. As shown in Fig. 3, the more close to the origin of coordinate the corresponding particle size parameters tend to infinitely small. To attention that the inversion results are sensitive to negative refraction index of particles, however, the stochastic algorithm can solve for

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multi-parameter, so such effects can be eliminated as long as seeking the real and imaginary part of particle negative refraction index as two unknown quantities.

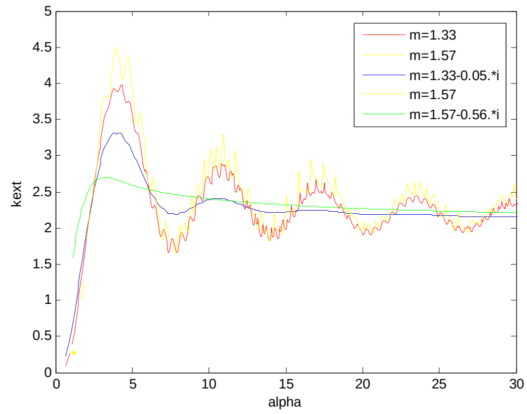

Figure 3. Relationship between extinction coefficient K_{ext} and α

As shown in Fig. 3, for the non-dissipative particles, the curve shocks obviously, there are glitches between a series of maximum and minimum values. The extinction coefficient tends to 2 as α value increases. For the dissipative particles which complex refractive index is plural, the shake of the whole curve reduced significantly and the phenomenon of glitches disappeared. Therefore, for the non-dissipative particles, the function curves of K_{ext} and α under different m almost show the same trend.

To discuss the accuracy of π_L, τ_L in the premise of a_L, b_L calculated accuracy is given. For the particle scattering is mainly decided by its size and refractive index, in MATLAB programming the refractive index is set as m=1.33-0.05.*i, Fig. 4, Fig. 5, Fig. 6, Fig. 7 show the relationship of scattering angle function and scattering angle in the case of particle size parameter α set as 5, 10, 15, 20.

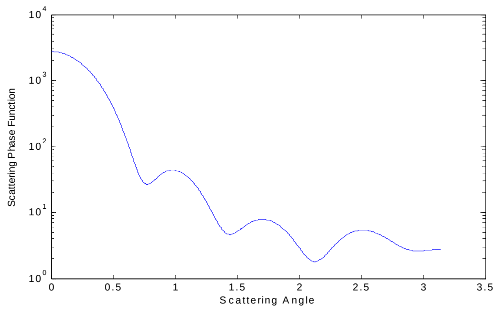

Figure 4. Scattering angle function distribution when $\alpha = 5$

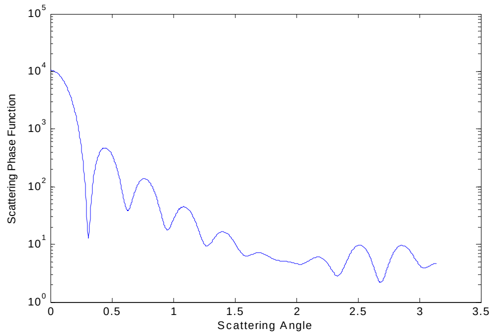

Figure 5. Scattering angle function distribution when $\alpha = 10$

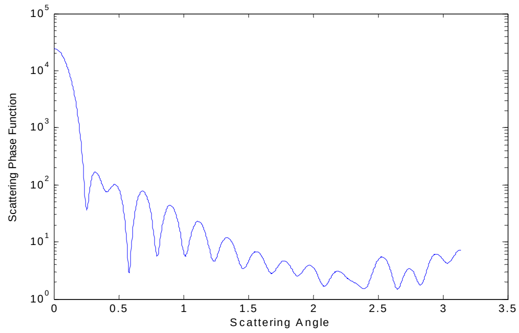

Figure 6. Scattering angle function distribution when $\alpha = 15$

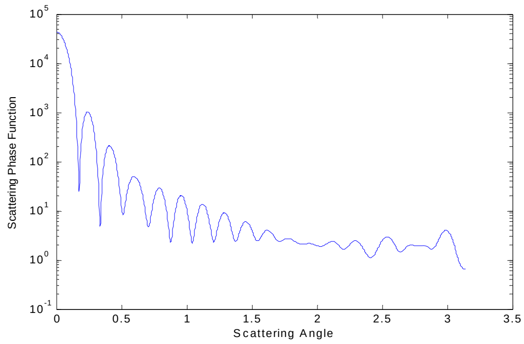

Figure 7. Scattering angle function distribution when $\alpha = 20$

Another key of the numerical calculation of Mie theory is the rational selection of the upper limit frequency L. The convergence problem discussed in this paper primarily focuses on the convergence number, to assume

$$L = 2\,\mathrm{Re}(m)\alpha + 10 \qquad (27)$$

Here m is particle refractive index, α is particle size parameter. Thus, the cycle index of program is fixed basically, and then to minimize the sum number. The criteria of judging convergence is: Until the stable output of ten digits after the decimal point. It can be found through comparison that the series summation number of

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this algorithm is less than the upper limit frequency of other algorithms. The calculation speed will be faster in the same case.

IV. Inversion Algorithm

Using white light as incident light source, scattering spectrum $I(\lambda_i)$ can be measured by monochromator, and combining with $K(\theta,\lambda,a)$ obtained by MATLAB programming, particle size distribution $f(a)$ can be solved through formula1. However, the direct solution for integral inversion in formula is very difficult, so a random algorithm, Markov chain will be used to inverse in this paper.

There is a random process $s(t)$, $t\in T$, T is a fixed set of real numbers, if $t_0 < t_1 < \cdots < t_n$ for random n moments in the process, when $s(t_0), s(t_1),\cdots, s(t_{n-1})$ is given, the condition distribution of $s(t_n)$ only depends on the nearest known value $s(t_{n-1})$:

$$P(S_n = s_n | S_{n-1} = s_{n-1},\cdots,S_1 = s_1, S_0 = s_0) = P(S_n = s_n | S_{n-1}) \quad (28)$$

Therefore, Markov chain is no aftereffect, the future is independent with the past under known current conditions.

For simplicity, to assume the total number of particles is N, the frequency distribution of particles is f_r, then $f_r \in M_r$, $NM_r \in M_n$, so the approximate solution $f_{r,0}$ should satisfy

$$\rho_\Omega(NKf_{r,0}, I) = \inf_{f_r \in M_r} \rho_\Omega(NKf_r, I) \quad (29)$$

Here, $f = (y_1, y_2, \cdots, y_n)$, $f_r = (y_{r1}, y_{r2}, \cdots, y_{rn})$. On the other hand,

$$\rho_\Omega(NKf_r, I) = \left[\sum_{i=1}^{w}\left(\sum_{j=1}^{n} NA_{i,j} y_{r,j} - I_i\right)^2\right]^{1/2} \quad (30)$$

Unknown value N is determined by following formula

$$N = \frac{1}{w}\sum_{i=1}^{w}\left(I_i \Big/ \sum_{j-1}^{n} A_{i,j} y_j\right) \quad (31)$$

Currently the analysis object and model structure are complicated in many application problems, so the large numbers of difficulties which cannot be achieved by deterministic algorithm that can be solved by using Markov chain to take a sample of Gaussian distribution, Rayleigh distribution, Jhonson-SB distribution or some other discrete distributions with large sample space and to do some random simulations. It is a dynamic Monte Carlo method that called Markov Chain Carlo method short for MCMC.

MCMC is a simple and effective method which has a widespread application in the areas of the significant testing and maximum likelihood estimate. The basic idea of MCMC is: To obtain the sample of m(x) through the establishment of the Markov chain with a stable distribution m(x), thus applied in all kinds of probability statistics. Therefore the stationary distribution is defined as that the Markov chain using P(x, dx) for its transition probability[7][8].

The process of solving the distribution function of particle size $f(a)$, in fact, is an approximation using limited（N） discrete y_j （j=1,2,3…..n）. If the particle size can be seemed as one by one state, then y_j could be regarded as y particles are accumulated in state j. Considering the Markov chain composed by these states, the function of particle size distribution can be solved through the views of probability theory as this: To assume P particles keeping doing random independent movement in Markov chain, and the moving direction of any particle x headed toward the direction reducing the criterion $\varphi(f_r)$. $\varphi(f_r)$ is defined as:

$$\varphi(f_r) = \sum_{i=1}^{W}[C\sum_{j=1}^{M} p_j^{(i)} J_{j,i} / P - I(\lambda_i)]^2 / I^2(\lambda_i) \quad (31)$$

Here, $\varphi(f_r)$ is the number of particles for any state j in initial moment i. The movement will stop when the moving number of $\varphi(f_r)$ reaches the set value, at this moment the particle size distribution is reflected by the number of all states particles. It is simple to prove the above Markov chain is ergodic and recurrence, so each particle is possible to reach any state, which guarantees the global convergence of the inversion algorithm.

V. Numerical Simulation

The range of wavelength is set from 300nm to 800nm, step length is 20nm, assuming that the particle refractive index m=1.57-0.56i is invariable, the scattering angle $\theta = 1^\circ$, selecting Jhonson-SB function as the model of particle size distribution.

Obviously, the measured data has a certain error, assuming the error is caused by the noise. In order to simulate the measured data, the researched particles are assumed to obey the distribution of Jhonson-SB function, $I(\lambda)$ can be calculated through integral, and then added the assumed signal to noise ratio. In this paper 5%, 10%, 20% are selected as the noise parameters, 1000 particles are picked in the inversion, the status number of Markov chain is 20 and the calculation will stop when movement number arrives at 20000. The parameters of Jhonson-SB function $\sigma = 1.8$, $\mu = 3.7$.

Fig. 8 shows the inversion results with the noise of 0, 5%, 10% and 20%. It can be seen from Fig. 8 that the

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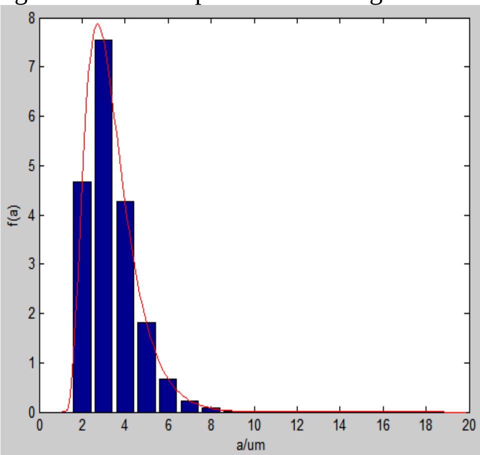
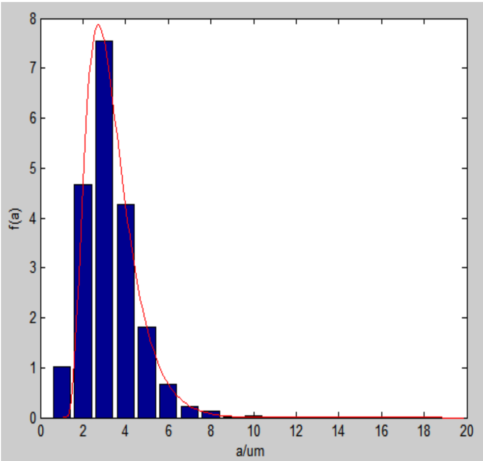
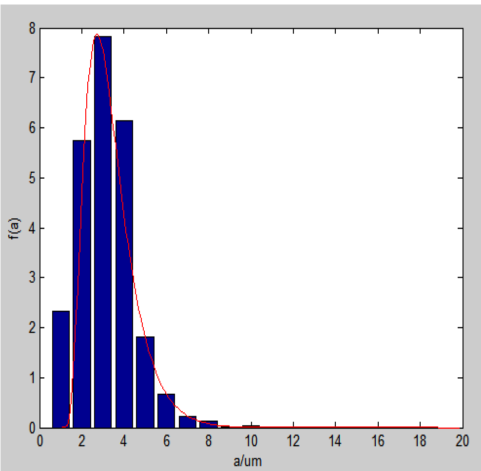
inversion result has a certain error in the range of small particle size, when the noise increases to 20%, the inversion result of particle size differs obviously in part of the particle size range. The results of stochastic inversion are reliable and noise immunity is still very good in the noise range 20%. Overall, it is very advantageous terms compared to other algorithms.



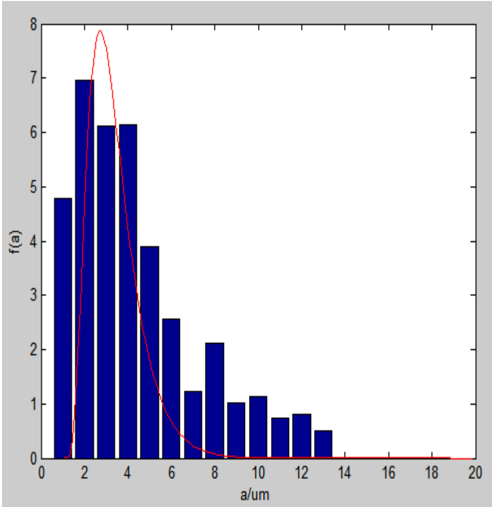
(a) Noise 0%



(b) Noise 5%



(c) Noise 10%



(d) Noise 20%

Figure 8. Inversion results of very narrow distribution with single peak

VI. Conclusion

Mie scattering theory is the basis of the particle size measurement based on light scattering method. In this paper, the expressions of the scattering parameters are given, and the improved algorithm of relative scattering coefficients of spherical particle is discussed. The built-in function module in MATLAB is fully utilized in programming. To observe from the simulation diagrams, the calculated results of these main scattering parameters are accurate, it has wide applicability and good convergence. The stochastic inversion algorithm applied in solving for particle size distribution has been researched in this paper, Markov chain algorithm is introduced and numerical simulation within an acceptable noise range is processed. It has strong anti-noise ability, and it is insensitive to the form of particle size distribution, so it does not rely on the priori information about relative measuring system. However, there are some shortages existed in stochastic inversion algorithm, it is sensitive to the complex refractive index of particles, and the calculation speed is slow compared to the deterministic algorithm. In any case, the stochastic inversion algorithm has a great research value that its superiority brings online measurement of particle size distribution very good prospects.

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