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ANALYSIS OF NON-NEGATIVE TIKHONOV AND TRUNCATED SINGULAR VALUE DECOMPOSITION REGULARIZATION INVERSION IN PCS

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ABSTRACT

Considering non-negative characteristic of the particle size distribution (PSD), based on trust-region-reflective Newton method, two non-negative regularization methods of truncated singular value decomposition (TSVD) and Tikhonov (TIK) for photon correlation spectroscopy (PCS) are proposed in this paper. Combining two regularization parameter criterions of GCV and L-curve, two non-negative regularization methods are studied. The study results show that, compared with TIK, TSVD has bigger truncation effect, poorer smoothness and narrower distribution width of inversion PSD, in the case of noise, TSVD has smaller relative error and peak value error of PSD, better capacity to discriminate bimodality and stronger anti-noise, but at noise-free case, TSVD hasn't obvious advantages, TIK and TSVD are respectively more suitable for using GCV and L-curve criterion to determine the regularization parameter.

Keywords: *Photon Correlation Spectroscopy, Particle Size, Non-negative Regularization, Inversion*

1. INTRODUCTION

Photon correlation spectroscopy (PCS, is also called dynamic light scattering) technology has become an effective method for measuring sub-micron and nano-particles size[1], which obtain the particle size distribution (PSD) by measuring and inverting the autocorrelation function (ACF) of the intensity fluctuations scattered by the investigated sample. However, inverting PSD from ACF is a high ill-posed problem, which has been the difficulty in PCS technology. Only when ACF measured is no noises and data calculation is no rounding errors, does the equation have a unique solution in theory. In the measurement, because of the presence of the noise and rounding errors, the existence, uniqueness and stability of solutions are difficult to guarantee. Therefore, in PCS technology, we can't solve true PSD, and only obtain its approximate value. For approximate solution, researchers have proposed numerous methods such as Cumulants method [2], exponential sampling method [3], the Bayesian strategies method [4], and the neural network method [5]. Considers non-negativity of PSD, there are some other methods such as CONTIN [6] and NNLS [7], which have been verified to improve the inversion accuracy of PSD. However, these methods have the drawbacks of sensitivity to noise, difficulty in choosing a regularization parameter, and complications in operation, poor the capacity to discriminate bimodality.

For the solving of ill-posed equation, regularization is one of most powerful approaches. In PCS inversion, considering together the regularization and non-negativity should be very effective. Trust-region-reflective Newton method [8] is a widely used non-negative constraint method which is included in the Matlab toolbox. Therefore, based on Trust-region-reflective Newton method, non-negative regularization for PCS is proposed in this paper. The regularization are many methods such as Tikhonov (TIK) regularization, truncated singular value decomposition (TSVD) regularization, landweber regularization, conjugate gradient(CG) regularization and so on. So far, there is not an optimal regularization method suitable for any problem. Therefore, the study of different regularization methods is very meaningful in PCS inversion. In this paper, TIK and TSVD were compared. In regularization method, selection of regularization parameter is critical. The generalized

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cross-validation (GCV) criterion [9] and L-curve criterion [10] are more widely used in practice. GCV and L-curve criterion are also compared in the PCS. By the comparison, we find out that characteristics and suitable regularization parameter criterion of each method, and draw a conclusion that TSVD has better capacity to discriminate bimodality and stronger anti-noise.

2. INVERSION PRINCIPLES OF PCS

For the light field of the Gaussian distribution, ACF of scattered light intensity is given by a Siegert relationship. For the polydisperse particles, the normalized ACF of scattered light intensity is expressed as

$$g(\tau)=\int_0^{\infty}G(\Gamma)\exp(-2\Gamma\tau)d\Gamma \quad \int_0^{\infty}G(\Gamma)d\Gamma=1 \qquad (1)$$

where τ is the sampling time, Γ is the decay width, $G(\Gamma)$ is normalized distribution function of the decay width. In Eq.(1), the relationship of decay width and the particle size is as follow,

$$\Gamma=Dq^2,\ q=\frac{4\pi n}{\lambda_0}\sin(\frac{\theta}{2}),\ D=\frac{k_B \mathrm{T}}{3\pi\eta d} \qquad (2)$$

where D is diffusion coefficient, q is the scattering wave vector, n is the refractive index of the solvent, λ_0 is the wavelength of the incident light in vacuum, θ is the scattering angle, k_{B} is the Boltzman constant, T is absolute temperature, η is solvent viscosity, and d is the diameter of equivalent spherical particles. Eq.(1) is a high ill-posed equation. In theory, we can invert $G(\Gamma)$ from measured $g(\tau)$, $G(\Gamma)$ is retrieval PSD.

3. REGULARIZATION INVERSION METHOD

In the practical solution, Eq.(1) is discretized as

$$Ax=b \qquad (3)$$

where elements of matrix b , x and A are $b_j=g(\tau_j)$, $x_i=G(\Gamma_i)$ and $a_{i,j}=\exp(-2\Gamma_i\tau_j)$, respectively. Solution of Eq.(3) can be expressed as the following least squares(LS) problem,

$$\|Ax-b\|_2^2=\min=x_{LS} \qquad (4)$$

A powerful tool for the solving of LS problem is the singular value decomposition (SVD). Then, SVD of A is as follows

$$A=U\Sigma V^T=\sum_{\mathrm{i=1}}^{\mathrm{n}}u_i\sigma_i v_i^T \qquad (5)$$

where $\Sigma=diag(\sigma_1,\sigma_2,\cdots\sigma_{\mathrm{n}})$, σ_i are the singular values of A, U and V is left and right singular values vectors, respectively.

LS solution of Eq. (3) is also expressed as

$$x_{LS}=\sum_{i=1}^{\mathrm{n}}\frac{<u_i,b>}{\sigma_i}v_i \qquad (6)$$

In Eq.(6), if b contains noises, then σ_i will be infinitely small, which will lead to big deviation of x_{LS}. For solving of this problem, regularization is an effective method.

3.1 Tikhonov Regularization (TIK)

TIK adds the 2-norm of solution as a constraints condition to Eq.(3). Then, LS solution of Eq.(3) is approximately equal to following problem.

$$\min\ \{\|Ax-b\|_2^2+\lambda^2\|L\ (x-x_0)\|_2^2\} \qquad (7)$$

where L is unit matrix, x_0 is initial solution, λ is the regularization parameter.

When $x_0=0$, based on SVD theory, TIK solution of Eq.(3) is can expressed as

$$x_{TIK}=\sum_{i=1}^{\mathrm{n}}(\frac{\sigma_i^2}{\sigma_i^2+\lambda^2})\frac{<u_i,b>}{\sigma_i}v_i \qquad (8)$$

From Eq.(8), we can see that TIK solution of Eq.(3) actually adds the following filter factors to the original solution of Eq.(3).

$$f_i=\frac{\sigma_i^2}{\sigma_i^2+\lambda^2} \qquad (9)$$

In order to avoid solving of SVD and reduce the computation, Eq.(8) can also be expressed as

$$\min\left\{\left\|\begin{bmatrix}A\\ \lambda I\end{bmatrix}x-\begin{bmatrix}b\\0\end{bmatrix}\right\|_2^2\right\} \qquad (10)$$

3.2 Truncated Singular Value Decomposition Regularization (TSVD)

Principle of TSVD is to remove small singular value which amplifies the disturbances. Principle of TSVD is to remove small singular value which amplifies the disturbances. Based on this principle, after truncating small singular value, matrix A can be expressed as

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$$A_k = \sum_{i=1}^{k} u_i \sigma_i v_i^T \qquad (11)$$

where k is regularization parameter.

Thus, solving of Eq.(3) is changed into solving of following well-posed equations

$$A_k x = b \qquad (12)$$

According to Eq.(5) and Eq.(11), Solution of Eq.(12) is expressed as

$$x_{TSVD} = \sum_{i=1}^{k} \frac{< u_i, b >}{\sigma_i} v_i \qquad (13)$$

From Eq.(13), we can see that filter factors of TSVD is

$$f_i = \begin{cases} 1 & \sigma_i > \lambda \\ 0 & \sigma_i < \lambda \end{cases} \qquad (14)$$

where λ is threshold, which meets $\sigma_1 > \sigma_2 > \cdots \sigma_k > \lambda > \sigma_{k+1} > \cdots \sigma_n > 0$

Solution of TSVD can also be expressed as

$$\min \| A_k x - b \|_2^2 \qquad (15)$$

3.3 Selection of the Regularization Parameter

3.3.1. L-curve criterion

L-curve criterion computes the curvature of the following curve in log-log scale $(lg(\|x_\lambda\|_2), lg(\|b - Ax_\lambda\|_2))$ (with λ as its parameter) and seek the point with maximum curvature, which is defined as the curve's corner. The curve's corner λ corresponds to the optimal regularization parameter value. For discrete ill-posed problems, because this curve almost always has a character-istic L-shaped appearance with a distinct corner separating the vertical and the horizontal parts of the curve. So hence its name is called as L-curve criterion.

3.3.2 Generalized cross-validation (GCV) criterion

Generalized cross-validation (GCV) selects the regularization parameter by minimizing the following GCV function

$$G = \frac{\| Ax_{reg} - b \|_2^2}{[trace(I - AA^I)]^2} \qquad (16)$$

where G is defined as regularization parameters , A^I is a matrix which produces the regularized solution x_{reg} when multiplied with b , i.e, $x_{reg} = A^I b$,Trace represents the matrix trace.

4. ANALYSIS OF SIMULATION DATA

The solving of Eq.(10) and Eq.(15) are achieved by trust-region-reflective Newton method. In Eq.(10) and Eq.(15), two criterions of regularization parameter are respectively used, and form four methods such as TIK+L-curve, TIK+GCV, TSVD+ L-curve and TSVD+ GCV. In order to verify the validity of non-negative TIK and TSVD based on trust-region-reflective Newton method and study their characteristic in the PCS inversion, using four methods, noise-free and noisy ACFs of unimodal and bimodal distributions particles were inverted. Their inversion results are shown in Figs.1~6, Inversion data of PSD are shown in Tables1~4. In the Tables1~4, the relative error is defined as

$$\text{relative error} = \| x - x_{theory} \|_2 / \| x_{theory} \|_2 \qquad (17)$$

In the study, noise-free and noisy ACFs with noise level of 0.005 and 0.01 respectively were acquired by the simulation. The simulation initial PSD is Johnson's SB distribution [11].

In the simulations, the unimodal distributions particles share parameters of Johnson's SB ρ =0, β =1.1, α_{max} =600nm and α_{min} =250nm, the bimodal distributions particles utilized the sum of two Johnson's SB functions of equal intensity quotients, sharing parameters ρ_1 =3.8, β_1 =2.1, ρ_2 =-2.4, β_2 =2.0, α_{max} =700nm and α_{min}=100nm. Simulation experiment conditions are as follows, the wavelength is 632.8nm, the refractive index of scattering medium(water) 1.331,scattering angle 90°, absolute temperature 25℃, Botlzman constant 1.3807×10^{-23} $J \cdot K^{-1}$,the viscosity coefficient of water 0.89×10^{-3} $N \cdot S \cdot K^{-1}$.

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Tabel.1 Inverted Data Of Unimodal Particles For TIK In The Different Criterion

noise levels	L-curve			GCV		
	peak value/nm	peak value error %	relative error	peak value/nm	peak value error %	relative error
0	400	5	0.1213	400	5	0.1213
0.005	410	2.5	0.2087	410	2.5	0.2087
0.001	410	2.5	0.6209	410	2.5	0.3209

Tabel.2 Inverted Data Of Unimodal Particles For TSVD In The Different Criterion

noise levels	L-curve			GCV		
	peak value/nm	peak value error %	relative error	peak value/nm	peak value error %	relative error
0	400	5	0.1246	400	5	0.1813
0.005	410	2.5	0.2034	410	2.5	0.2034
0.01	410	2.5	0.1784	410	2.5	0.2882

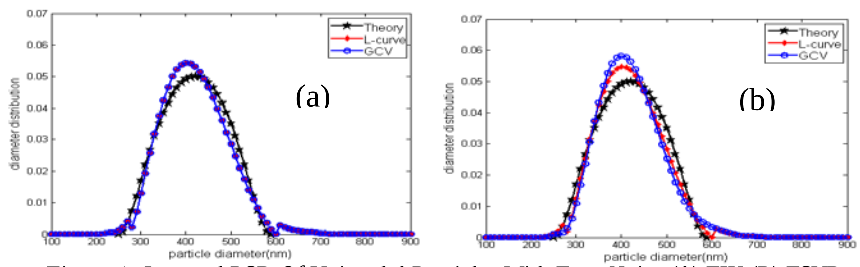

Figure.1: Inverted PSD Of Unimodal Particles With Free-Noise (A) TIK (B) TSVD

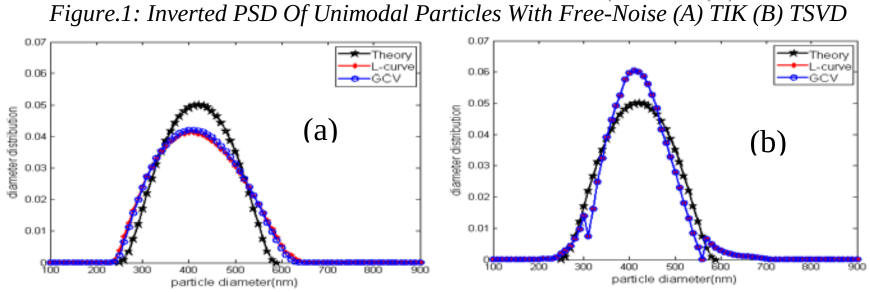

Figure.2: Inverted PSD Of Unimodal Particles At Noise Levels 0.005 (A) TIK (B) TSVD

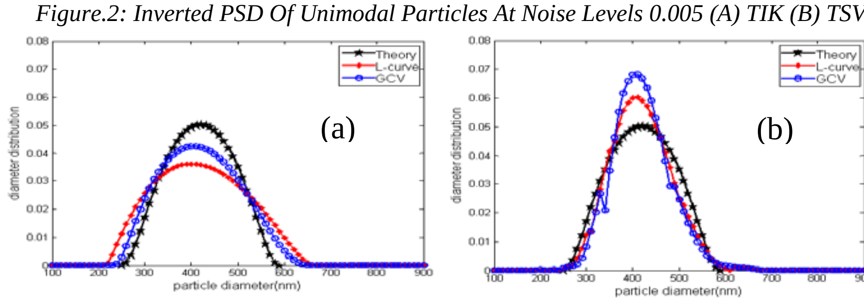

Figure.3: Inverted PSD Of Unimodal Particles At Noise Levels 0.01 (A) TIK (B) TSVD

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Tabel.3 Inverted data of bimodal particles for TIK in the different criterion

noise levels	L-curve			GCV		
	peak value/nm	peak value error %	relative error	peak value/nm	peak value error %	relative error
0	160.8，553.6	3.94, 3.48	0.4777	160.8，553.6	3.94, 3.48	0.4777
0.005	—	—	0.6239	160.8，540.3	3.94, 5.80	0.4378
0.01	—	—	0.7138	—	—	0.6249

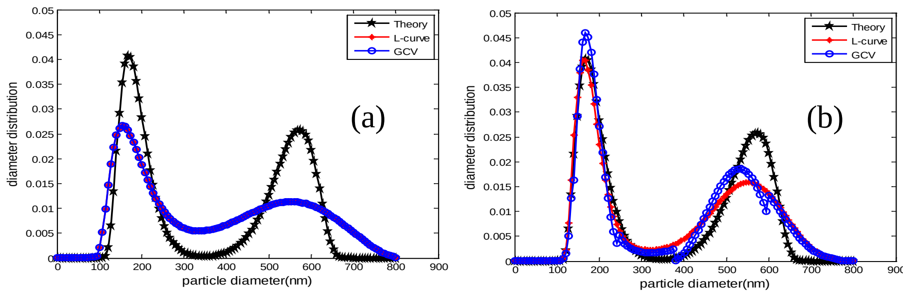

Figure.4: Inverted PSD Of Bimodal Particles With Free-Noise (A) TIK (B) TSVD

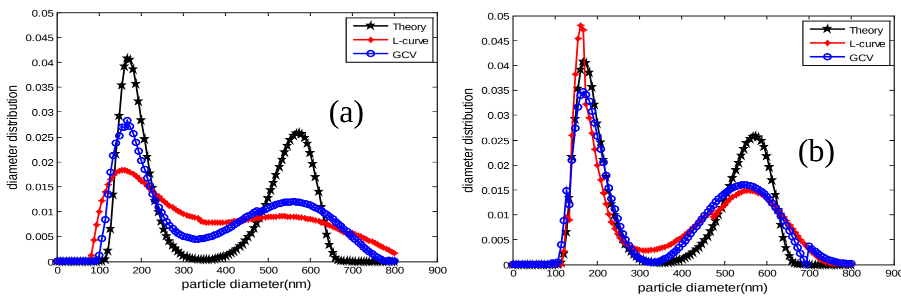

Figure.5: Inverted PSD of bimodal *Particles* at noise levels 0.005 (a) TIK (b) TSVD

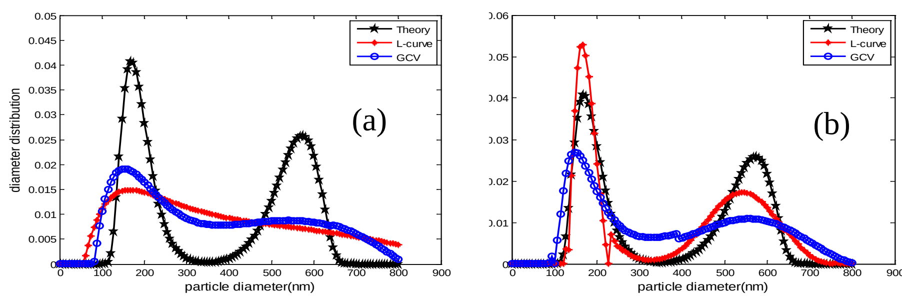

Figure.6: Inverted PSD Of Bimodal Particles At Noise Levels 0.01 (A) TIK (B) TSVD

Tabel.4 Inverted Data Of Bimodal Particles For TSVD In The Different Criterion

noise levels	L-curve			GCV		
	peak value/nm	peak value error %	relative error	peak value/nm	peak value error %	relative error
0	167.4, 553.6	0, 3.48	0.2681	167.4,533.6	0, 6.97	0.2808
0.005	160.8, 560.3	3.94, 2.31	0.3417	167.4,546.9	0, 4.65	0.2696
0.01	167.4, 533.6	0, 6.97	0.3402	154.1,553.6	7.94, 3.48	0.5130

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From theirs inversion results of the Figs 1~6 and Tables1~4, when non-negative constraints of TIK and TSVD were achieved by Trust-region-reflective Newton method, we can find out the following phenomenon and draw some conclusions.

1) For the unimodal distributions particles, inversion results of TIK and TSVD are the unimodal PSD. For the bimodal distributions particles, inversion results of TSVD are the bimodal PSD, at noise level 0~0.005, TIK +GCV can obtain the bimodal PSD, at noise level 0.01, PSD of TIK +GCV hasn't apparent bimodal characteristics. Therefore, at noise level 0~0.01, TSVD can use for inversion of the unimodal and bimodal distributions particles, while TIK can use for inversion of the unimodal distributions particles and bimodal distributions particles with noise level of less than 0.005.

2) The inversion results of TIK and TSVD may emerge fluctuations. Inversion PSD of TIK is relatively smoother, while that of TSVD fluctuates more serious. This is because different filter window for truncating singular value is respectively used in TIK and TSVD. Different filter window has different truncation effects, which leads to fluctuations of inversion PSD. Steeper the window function is, more outstanding truncation effect is. Taking inversion of unimodal distribution particles with noise level 0.005 as an example, filter windows of TIK and TSVD are shown in Fig.7. The singular value of TSVD was truncated by the rectangular window function in filtering process. Rectangular window function is steeper, its truncation effect is outstanding and fluctuations phenomenon of inversion PSD is more severe. Compared with TSVD, filtering window of TIK is smoother, its truncation effect is weaker, and its inversion PSD is relatively smoother.

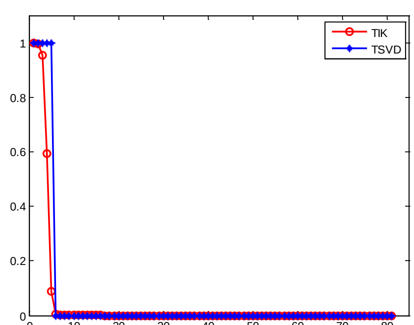

Figure.7: Filter Window Function Of TIK And TSVD

3) For a unimodal distributions particles inversion, peak value error of two methods is the same. However, from inversion relative error point of view, at free-noise case, relative error of TIK is smaller, at the noise case, compared with TIK, relative error of TSVD is significantly lower and is lower than that of TIK up to 0.4425; For the bimodal distribution particles inversion, compared with TIK, at all noises cases, relative errors of TSVD were lower and is lower than that of TIK up to 0.3736. From the peak value error the point of view, at noise-free case, peak value error of TSVD + GCV is 6.97%, it was significantly higher than that of TIK. However, at all noise cases, compared with TIK, peak value error of TSVD is smaller and most reduce the peak error 3.94%. From the capacity to discriminate bimodality the point of view, capacity to discriminate bimodality of TSVD is stronger than TIK, at noise level 0.01, inversion results of TSVD still has apparent bimodal characteristics, while inversion results of TIK hasn't apparent bimodal characteristics. From the peak width the point of view, in all noise cases, inverted PSD of TIK is wider than that of TSVD.

In summary, we can draw the following conclusions: in the noise cases, TSVD has smaller relative error and peak value error, better capacity to discriminate bimodality, a stronger anti-noise, narrower peak width, but at noise-free cases, TSVD haven't obvious advantage than TIK.

4) By comparing L-curve and GCV regularization parameter criterion in the inversion of TIK and TSVD, we can be seen that, for the unimodal distribution particles, peak value error of TIK +GCV and TIK +L-curve is same, but at the noise level 0.01, relative error of GCV criterion is significantly lower than that of L-curve, for the bimodal distribution particles, when the noise level is greater than or equal to 0.005, inversion result of L-curve criterion can not discriminate two peaks, while GCV criterion can clearly distinguish two peaks at the noise level lower than 0.01, at the noise level 0.01, inversion PSD of GCV criterion still has weak characteristic of two peaks. Therefore, the GCV criterion is more suitable to determine the regularization parameter for TIK in PCS inversion; when inverting PSD by TSVD, GCV and L-curve criterion can get more accurate inversion results, however, compared with GCV criterion, peak value error of L-curve criterion is smaller, maximum peak value error of TSVD+L-curve is less than 6.97%, while that of TSVD + GCV is 7.94%, moreover, relative error of TSVD + GCV is generally higher, at noise level 0.01, its capacity to discriminate two peaks is relatively poorer. Therefore, L-curve criterion is more suitable to determine the regularization parameter for TSVD in PCS inversion.

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5. EXPERIMENTAL RESULTS

ACF of scattered light intensity was obtained through PCS experiment setup of our research group[12].The materials tested were standard polystyrene latex spheres suspended in purified water. They are unimodal particle with average diameters 100nm and bimodal distributions particle with average diameters 60 nm and 200 nm , For the latter, the relative proportion of two samples was approximately 1:1. All measurements were made at scattering angle of 90° and temperture 298 2 K.

Using TIK+L-curve, TIK+GCV, TSVD+L-curve and TSVD+GCV, respectively, the above measured ACF data were inverted. The inversion PSDs and data are shown in Figs.8 ~9 and Table 5.

As can be noted from Figs.8~9 and Table5, inversion PSD of TIK and TSVD are both consistent with their true PSD. However, compared with TIK, the peak value of TSVD is more close to the true value, its peak width is also narrower. It shows that TSVD has good noise immunity. But inversion PSD smoothness of TSVD is worse. In terms of regularization parameters criterion, inversion results of TIK + GCV and TSVD + L-curve more close to true distribution. Therefore, from the analysis of the measured data, we can draw the conclusion consistent with simulation data.

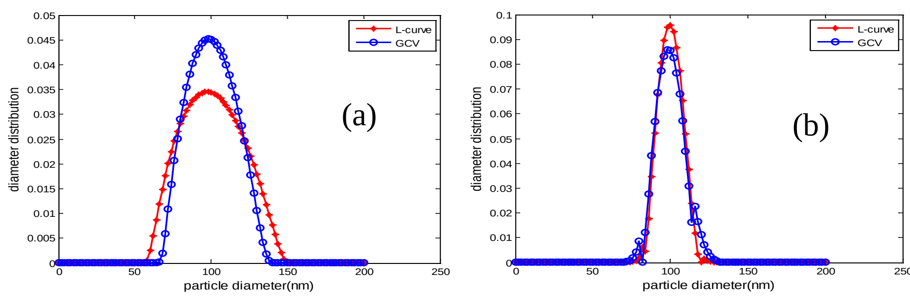

Figure.8: Inversion PSD Of 100nm Particles (A) TIK (B) TSVD

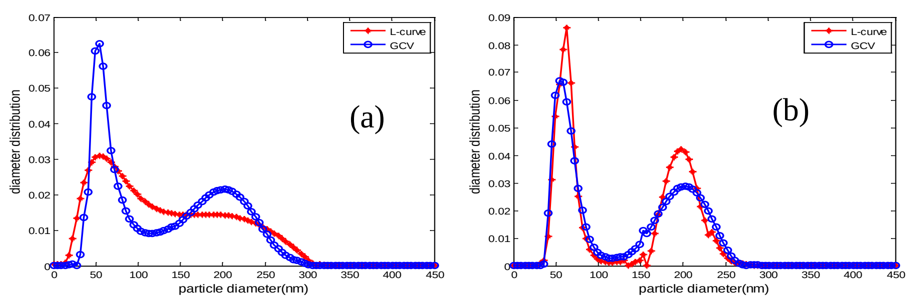

Figure.9: Inversion PSD Of 60nm And 200nm Particles (A) TIK (B) TSVD

Table 5 Inversion Peak Value Of Experimental Unimodal And Bimodal Particles

Particles (nm)	TIK		TSVD	
	L-curve	GCV	L-curve	GCV
100	98	98	100	100
60，200	—	54, 202.5	63, 198	54,202.5

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6. CONCLUSION

For inversion problem of PCS, based on trust-region-reflective Newton method, at regularization parameter criterion of GCV and L-curve, two non-negative constraints regularizations with TIK and TSVD were proposed and compared in this paper. By the study of simulation and experiment data of unimodal and bimodal distributions particles, we can be drawn the following conclusions. Firstly, compared with TIK, truncation error of TSVD is bigger, smoothness and peak width of its inversion PSD is poorer and narrower, respectively. Secondly, in the case of noise, inversion PSD of TSVD has smaller relative error, smaller peak value error, better capacity to discriminate bimodality and stronger anti-noise, but noise-free case, TSVD hasn't obvious advantages. Thirdly, for PCS inversion, TIK and TSVD are respectively more suitable for using GCV and L-curve criterion to determine the regularization parameter. These conclusions can be treated as a reference for the application of regularization method in PCS inversion.

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REFERENCES:

[1] K. Takahashi, H.Kato and T.Saito, "Precise measurement of the size of nanoparticles by dynamic light scattering with Uncertainty Analysis," Part. Part. Syst. Charact, Vol. 25, No. 1, 2008, pp. 31-38.
[2] D. E. Kopple, "Analysis of macromolecular polydispersity in intensity correlation spectroscopy:the method of cumulants," J. Chem. Phys, Vol.57, No. 11,1972, pp. 4814-4820.
[3] N. Ostrowsky, D. Sornette, P. Parker, and E. R. Pike, "Exponential sampling method for light scattering polydispersity analysis," J. Mod. Opt, Vol. 28, No. 8, 1981, pp. 1059-1070.
[4] M. Iqbal, "On photon correlation measurements of colloidal size distributions using Bayesian strategies," J. Comput. Appl. Math, Vol. 126, No. 1-2, 2000, pp.77-89.
[5] L. M Gugliotta, G. S Stegmayer and L.A Clementi, "A neural network model for estimating the particle size distribution of dilute latex from multiangle dynamic light scattering measurements," Part Part Syst Charact, Vol. 26, No.4, 2009, pp.41-52.
[6] S .W. Provencher, "CONTIN: A general purpose constrained regularization program for inverting noisy linear algebraic and integral equations," Comput Phy Commun, Vol. 27, 1982, pp.229-242.
[7] I. D. Morrison and E. F. Grabowski, "Improved Techniques for Particle Size Determination by Quasi-Elastic Light Scattering," Langmuir, No. 1, 1985, pp.496-501
[8] Coleman, T.F. and Y. Li, "An Interior, Trust Region Approach for Nonlinear Minimization Subject to Bounds," SIAM Journal on Optimization, Vol. 6, No. 2, 1996, pp.418-445.
[9] G.H.Golub, M.T.Heath and G. Wahba. "Generalized cross-validation as a method for choosing a good ridge parameter," Technometrics, Vol.21, 1979, pp.215-223.
[10] P. C. Hansen. "The Use of the L-Curve in the Regularization of Discrete Ill-posed Problems," SIAM J Sci Comput, Vol. 14, No.6, 1993, pp.1487-1503.
[11] A. B .Yu, N.Standish. "A study of particles size distribution," Powder Technology, Vol. 62, 1990, pp. 101-118.
[12] W. Liu, J. Shen, and X. Sun, " Design of multiple-tau photon correlation system implemented by FPGA". Proceedings of the International Conference on Embedded Software and Systems, Inst. of Elec. and Elec. Eng. Computer Society, July, 29-31, 2008, pp. 410-414.