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Non-negative Tikhonov Regularization Inversion Combining Trust-region with Interior Point Newton for Photon Correlation Spectroscopy

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Abstract: In order to improve the accuracy of inversion Particle Size Distribution (PSD) in the Photon Correlation Spectroscopy (PCS) technology, considering non-negative characteristic of PSD, based on Tikhonov regularization method, two non-negative constraint methods of trust-region (Trust) and Interior Point Newton (IPN) are compared in this study. Combining characteristics of two methods, an inversion method of Trust-IPN-Tikhonov is proposed. This method inherits the advantages of the Trust-Tikhonov and IPN-Tikhonov. The inversion results of simulation data and experimental data demonstrate that Trust-IPN-Tikhonov has smaller peak error, relative error and narrower distribution width than IPN-Tikhonov, compared with the Trust-Tikhonov, Trust-IPN-Tikhonov has not only smaller peak error and relative error but also better smoothness. All in all, Trust-IPN-Tikhonov has higher accuracy, better smoothness and is more consistent with the true distribution.

Key words: Photon correlation spectroscopy, inversion, tikhonov regularization, trust-region, interior point newton

INTRODUCTION

Photon correlation spectroscopy (PCS, is also called dynamic light scattering) technology has become an effective method for measuring sub-micron and nano-particle size (Cheng *et al*., 2009) which measures the time-averaged Autocorrelation Function (ACF) of the intensity fluctuations scattered by the investigated sample and obtain the information on Particle Size Distribution (PSD). However, obtaining PSD from measured ACF needs invert a first-kind Fredholm integration equation which is a high ill-posedness. The measurement data with small noises causes large deviation or instability of the solution. Therefore, solving of PSD becomes the difficulty of PCS technology. For this problem, numerous approaches have been proposed, such as Cumulants method (Koppel, 1972), CONTIN method (Provencher, 1982), NNLS method (Morrison *et al*., 1985) and more recently developed the neural network approach (Gugliotta *et al*., 2009) and the genetic algorithm (Li, 2008). However, these methods have some limitations of sensitivity to noise and lower accuracy. So far, the inverse problem is still current research focus. In practice, for solving of ill-posed equation, Tikhonov regularization method is one of most effective approaches (Liu, 2005). So, Tikhonov regularization method may be use for PCS inversion. To further improve inversion accuracy, considering non-negativity of PSD, the solution is imposed by non-negative constraint in the solution process. The solution will be limited to a relatively small area. Accordingly, inversion PSD can be improved (Roig and Alessandrini, 2006). In mathematics, non-negative constraint methods have numerous. Among them, the Trust-region (Coleman and Li, 1996) is a non-negative constraint method which is widely used for the field of engineering. When this method is used for Tikhonov regularization inversion, relative error of its inversion PSD is smaller. However, its smoothness is sometimes worse (Wang *et al*., 2012). Interior Point Newton method (IPN) (Bellavia *et al*., 2006) is also a non-negative constraint method. When this method uses for the PCS inversion, its inversion PSD has good smoothness, but its relative error is larger. Integrating the characteristics of the two methods, in PCS technology, a non-negative Tikhonov regularization inversion combining trust-region with interior point Newton (Trust-IPN-Tikhonov) is proposed in this study. Inversion results of this method have higher accuracy and better smoothness.

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TIKHONOV REGULARIZATION 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:

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

where, τ is the sampling time, $\Gamma$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:

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

where is diffusion coefficient, q is the scattering wave vector, nis 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. In theory, we can invert $G(\Gamma)$ from measured $g(\tau)$, $G(\Gamma)$ is retrieval PSD. In the practical solution, Eq. 1 is discretized as:

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

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. Tikhonov regularization solves least squares solution of Eq. 3 by following minimum problem.

$$\min\left\{\|ax - b\|_2^2 + \lambda^2\|(Lx - x_0)\|_2^2\right\} \quad (4)$$

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

When $x_0 = 0$, considering the non- negativity of PSD, Eq. 4 can also be changed into the following LS problem.

$$\begin{cases} \min\left\{\left\|\begin{bmatrix} a \\ \lambda L \end{bmatrix} x - \begin{bmatrix} b \\ 0 \end{bmatrix}\right\|_2^2\right\} = \min f(x) = \|Ax - B\|_2^2 \\ \text{st} \quad x \geq 0 \end{cases} \quad (5)$$

Choosing the appropriate regularization parameter, the PSD can be solved. The choice of regularization parameter uses Generalized Cross-Validation (GCV) criterion (Golub *et al*., 1979) in this study. This criterion can be expressed as:

$$\lambda = \frac{\|ax_{reg} - b\|_2^2}{[\text{trace}(I - AA^I)]^2} \quad (6)$$

where, a^I is a matrix which satisfies $x_{reg} = a^I b$, x_{reg} is regularized solution and trace represents the matrix trace.

NON-NEGATIVE TIKHONOV REGULARIZATION INVERSION COMBINING TRUST-REGION WITH IPN

Non-negative Tikhonov regularization inversion with Trust-region (Trust-Tikhonov): Trust-region (Coleman and Li, 1996) is a large-scale bound constrained optimization algorithm which uses trust region and interior point methods to solve the boundary constrained quadratic programming problems of Eq. 7 or 8:

$$\min_{x>0}\left\{f(x) = c^T x + \frac{1}{2}x^T Hx : 1 < x < u\right\} \quad (7)$$

$$\min_{x>0}\left\{\|Ax - B\| :\ 1 < x < u\right\} \quad (8)$$

Relation of Eq. 7 and 8 is $H = A^T A$, $c = -A^T Y$. Assuming $g(x) = \nabla f(x) = Hx + c$, defining $D = D(x)$, $D(x)$ is a diagonal matrix with i^{th} diagonal component equal to $|v_i(x)|^{1/2}$.

The local solution to Eq. 8 is the solution of the nonlinear system:

$$D^2(x)g(x) = 0 \quad (9)$$

Assuming $x^*$is the local minimum value of Eq. 7 and Eq. 8, if a feasible point x is sufficiently close to x^*, then the Newton step $S^N(x)$ can be defined as following system:

$$S^N(x) = -(D^2 H + JD^g)^{-1} D^2 g \quad (10)$$

where, $D^g = D^g(x) = \text{diag}(|g|)$. Each diagonal element of the diagonal Jacobian matrix J is defined as follows:

$$J_{ii} = \begin{cases} 1 & v_i = x_i - u_i \ \text{ or } \ v_i = x_i - l_i \\ 0 & \text{otherwise} \end{cases}$$

In each iteration, we maintain strict feasibility $1 < x^k < u$, S_k^N can be solve from following equation:

$$\bar{M}_k \bar{S}_k = -D_k g_k \quad (11)$$

where, $\bar{M}_k = D_k H_k D_k + J_k D_k^g$

Setting $s_k = D_k \bar{s}_k$, then according to the following iteration rule, we can estimate x:

$$x^{k+1} = x^k + \alpha_k S_k^N \tag{12}$$

where, $\alpha_k = (\beta_k^N)_{min} - \varepsilon_k$, $\varepsilon_k \le \chi_\alpha \|D_k g_k\|$, χ_α is a positive constant.

The above procedure is the approach of finding a local minimum. Using two-dimensional subspace decomposition technique, this approach can be extended to a global method and global optimal value can be solved. Specific process can be found in the literature (Coleman and Li, 1996).

Non-negative Tikhonov regularization inversion with IPN (IPN-Tikhonov): IPN method (Bellavia *et al*., 2006) solves the NNLS problem by searching the solution satisfying the Karush-Kuhn-Tucker (KKT) conditions. The solution of Eq. 5 is equivalent to the non-negative solution of the following nonlinear equations:

$$D(x)g(x) = 0 \tag{13}$$

where:

$$g(x) = \nabla f(x) = A^T(Ax - B),\ D(x) = \mathrm{diag}(d_1(x), \cdots d_n(x)),\ x \ge 0$$

$$d_i(x) = \begin{cases} x_i & g_i(x) \ge 0 \\ 1 & \text{otherwise} \end{cases}$$

Applying the Newton method to Eq. 13, the kth iterative step can be expressed as:

$$(D_k(x)A^TA + E_k(x))p = -D_k(x)g_k(x) \tag{14}$$

where, $E_k(x) = \mathrm{diag}\{e_1(x), \cdots e_n(x)\}$:

$$e_i(x) = \begin{cases} g_i(x) & 0 \le g_i(x) < x_i^s \ \text{ or } \ g_i(x)^s > x_i \\ 0 & \text{otherwise} \end{cases}$$

for $1 < s \le 2$.

If the solution is degenerate, the matrix $D_k(x)A^TA + E_k(x)$ may be singular. To avoid this case, Eq. 14 can be changed into the following form:

$$W_k(x)D_k(x)M_k(x)p = -W_k(x)D_k(x)g_k(x) \tag{15}$$

Where:

$$M_k(x) = A^TA + D_k(x)^{-1}E_k(x),\ W_k(x) = \mathrm{diag}\{w_1(x), \cdots w_n(x)\}$$

$$w_i(x) = (d_j(x), \cdots e_j(x))^{-1}$$

for x>0.

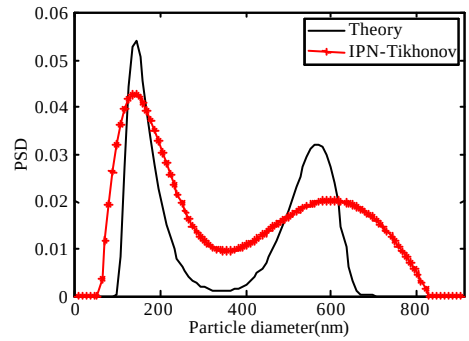

Fig. 1: Inversion PSD of IPN-Tikhonov

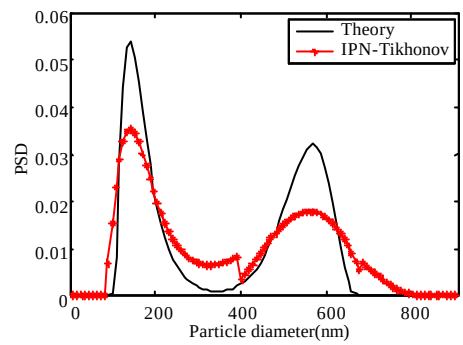

Fig. 2: Inversion PSD of Trust-Tikhonov

Table 1: Inversion data of IPN-Tikhonov and Trust-Tikhonov

Method	Peak value/nm	Peak value error (%)	Relative error
IPN- Tikhonov	143.5, 597.4	0, 4.67	0.6199
Trust- Tikhonov	143.5, 561.8	0, 1.56	0.3861

Equation 15 can be solved by LSQR method (Paige and Saunders, 1982). Specific process can be found in the literature (Bellavia *et al*., 2006).

In order to verify the effect of the above two non-negative methods, simulation ACF of bimodal distribution particle with noise level 0.001 is inverted by two methods. The simulation initial distribution is Johnson's SB function (Yu and Standish, 1990). The corresponding parameters are shown as follows: $u_1 = -2.2$, $\sigma_1 = 1.9$, $u_2 = 3.2$, $\sigma_2 = 1.5$, α_{max} =700 nm and α_{min} = 100 nm.The simulation experiment condition is as follows: the wavelength of incident beam is 632.8nm, the refractive index of scattering medium is 1.331, scattering angle is 90°, temperature is 25?,Botlzman constant is 1.3807×10^{-23} $J\bullet K^{-1}$ and the viscosity coefficient of water is 0.89×10^{-3} $N\bullet S\bullet K^{-1}$. When the inversion initial value is 0, the inversion results of above two methods are shown in Fig. 1 and 2. In the table, relative error $= \|x - x_{theory}\|_2 / \|x_{theory}\|_2$

From Fig. 1 and 2 and Table 1, we can see that, compared to IPN, inversion PSD smoothness of Trust is

poorer. But the peak value error and relative error of its inversion PSD are smaller and its inversion PSD is the closer to the theoretical distribution. On the contrary, using IPN, the peak value error and relative error of its inversion PSD are bigger. Besides, broadening of two modals of its inversion PSD is more serious. It means that inversion PSD of IPN is poorly agrees with theoretical distribution. However, inversion PSD smoothness of IPN is better.

Trust-IPN-Tikhonov inversion: For PCS inversion, the inversion PSD should have high accuracy and good smoothness. From the above analysis, we can know that inversion PSD of Trust has higher accuracy and poorer smoothness, while inversion PSD of IPN has better smoothness and poorer accuracy. Therefore, in order to get better inversion PSD, we combine the advantages of the Trust and IPN. A hybrid Trust-IPN-Tikhonov which combines Trust with IPN is proposed in this study. Firstly, this method inverts the PSD by using IPN-Tikhonov, then, the inversion result of IPN-Tikhonov is as the initial value and uses for inversion of Trust-Tikhonov. Thus, the inversion of Trust-Tikhonov is equivalent to the optimization near the optimal value. Accordingly, smooth optimal solution can easily be obtained.

In order to verify inversion results of Trust-IPN-Tikhonov, using the Trust-Tikhonov, IPN-Tikhonov and Trust-IPN-Tikhonov, respectively, simulation ACF with noise levels 0.001 and 0.005 were inverted. Simulation particles are unimodal distribution particle of 200~550 nm and bimodal distribution particle of 50~600 nm. Among them, the unimodal particles share parameters of Johnson's SB $u = 0.5$, $\sigma = 1.1$, $\alpha_{max} = 550$ nm and $\alpha_{max} = 200$ nm, the bimodal particles utilized the sum of two Johnson's SB functions of equal intensity quotients, sharing parameters $u_1 = 3.6$, $\sigma_1 = 2.0$, $u_2 = -2.3$, $\sigma_2 = 1.9$, $\alpha_{max} = 600$ nm and $\alpha_{min} = 50$ nm. Simulation experiment conditions are same as above. In the inversion, the inversion initial values of three methods are 0. The inversion results and data of three methods are shown in Fig. 3-4 and Table 2, 3.

Table 2: Inversion data of unimodal particles in the different noise levels

	Trust-Tikhonov			IPN-Tikhonov			Trust-IPN-Tikhonov		
Noise levels	Peak Value/nm	Peak value error %	Relative error	Peak Value/nm	Peak value error %	Relative Error	Peak Value/nm	Peak value Error %	Relative Error
0.001	322.86	4.15	0.1214	322.86	4.15	0.5209	318.57	2.76	0.0806
0.005	318.57	2.76	0.1539	331.43	6.91	0.5945	318.57	2.76	0.0826

Table 3: Inversion data of bimodal particles in the different noise levels

	Trust-Tikhonov			IPN-Tikhonov			Trust-IPN-Tikhonov		
Noise levels	Peak Value/nm	Peak value error %	Relative error	Peak Value/nm	Peak value error %	Relative Error	Peak Value/nm	Peak value Error %	Relative Error
0.001	110.84	4.57	0.2714	110.84	4.57	0.5434	116.15	0	0.2577
	477.07	2.18		503.61	3.26		477.07	2.18	
0.005	116.15	0	0.2116	116.15	0	0.5401	116.15	0	0.2004
	461.15	5.44		455.84	6.53		461.15	5.44	

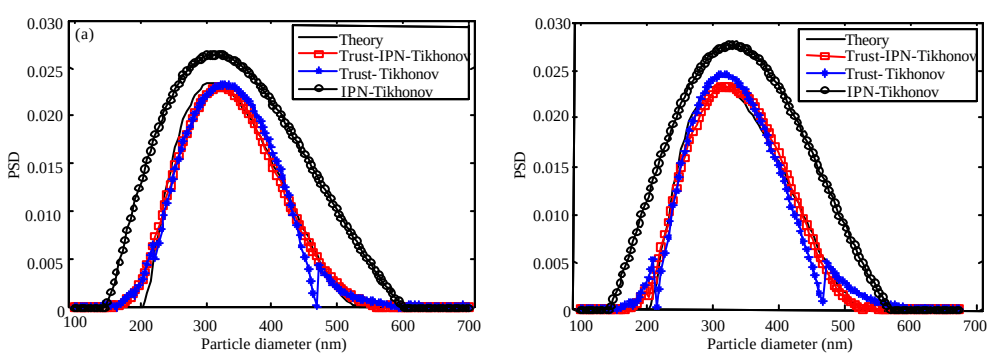

Fig. 3(a-b): Inversion PSD of unimodal particles at different noise levels (a) 0.001 and (b) 0.005

As shown in Fig. 3-4, Table 2, 3, compared with the Trust-Tikhonov, Trust-IPN-Tikhonov can reduce the peak error, relative error of inversion PSD in different degrees, unimodal distribution particle of 200~550 nm and bimodal distribution particle of 50~600 nm at most can improve the peak error and relative error of 1.39%, 0.0713 and 4.57%, 0.0137, respectively, besides, smoothness of its inversion PSD has clearly improved, it means that the inversion PSD of Trust-IPN-Tikhonov is better agreement with the theoretical PSD. Compared with the IPN-Tikhonov, for unimodal distribution particle of 200~550nm and bimodal distribution particle of 50~600nm, Trust-IPN- Tikhonov can improve the peak value error and the relative error of inversion PSD of 4.15%, 0.5119 and 4.57%, 0.3397, respectively and its inversion PSD is significantly narrower and more agree with the theoretical PSD. Therefore, we can observe that Trust-IPN-Tikhonov can combine the advantages of Trust-Tikhonov and IPN-Tikhonov to invert PSD. At the noise level with 0.001and 0.005, Trust-IPN-Tikhonov can get inversion results which are more agree with the theory distribution.

EXPERIMENT DATA INVERSION

ACF of measured particles is obtained by PCS system, the sample particles are the standard polystyrene latex particles which are unimodal distribution particles with average particle diameter 300 nm and bimodal distribution particles with average particle diameter 60 nm and 300 nm, scattering angle is 90°, experiment medium is water, the experiment temperature is 25°C. The measured ACF is inverted by above three methods. The inversion results and data are shown in Fig. 5 and Table 4. The inversion initial values of three methods are 0.

From Fig. 5 and Table 4, we can be seen, for unimodal distribution particle, the peak value errors of Trust-Tikhonov and Trust-IPN-Tikhonov are 1.03%, but the smoothness of Trust-IPN-Tikhonov is obviously superior to that of Trust-Tikhonov, for the bimodal distribution of particles, compared with the Trust-Tikhonov, inversion PSD of Trust-IPN-Tikhonov has better smoothness and its peak value error improves 1.28%. Compared with IPN-Tikhonov, for unimodal and

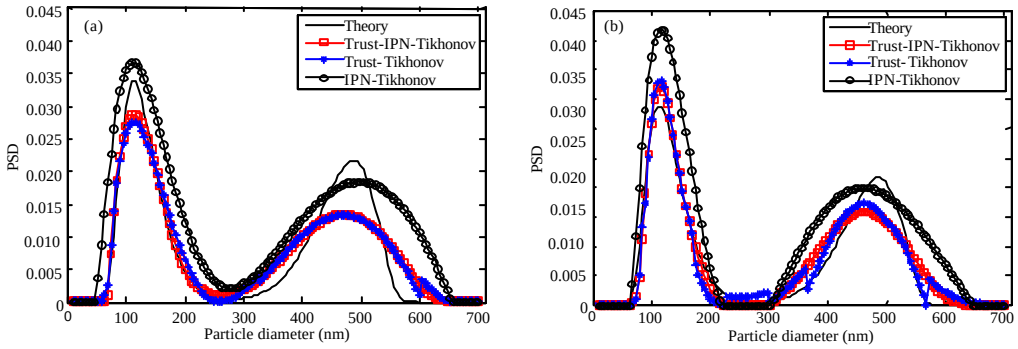

Fig. 4(a-b): Inversion PSD of bimodal particles at different noise levels (a) 0.001 (b) 0.005

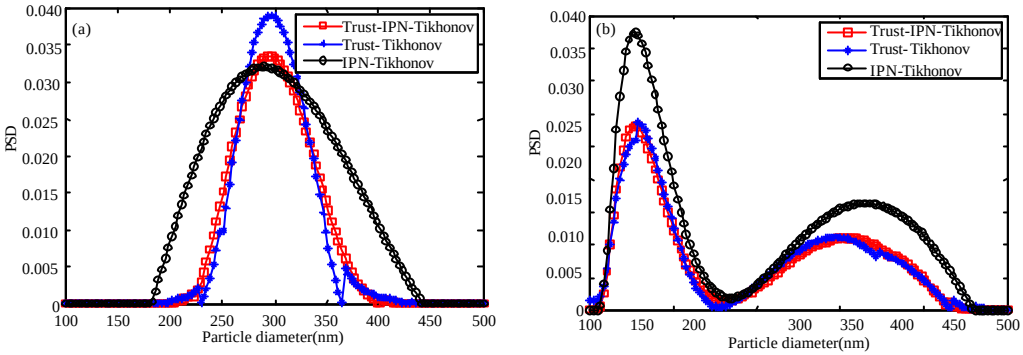

Fig. 5(a-b): Inversion PSD of experiment particles (a) 300 nm and (b) 60 and 300 nm

Table 4: Inversion data of experiment particles

	300 nm		60 and 300 nm	
Method	Peak value/nm	Peak value error (%)	Peak value/nm	Peak value error (%)
Trust- Tikhonov	296.92	1.03	58.58, 300.40	2.37, 0.13
IPN- Tikhonov	290.76	3.08	54.74, 327.27	8.77,9.09
Trust-IPN- Tikhonov	296.92	1.03	58.58, 304.24	2.37, 1.41

bimodal distribution particle, peak value errors of Trust-IPN-Tikhonov's inversion PSD is smaller and improve by 2.06 and 7.68%, respectively. Therefore, from the analysis of the measured particles, we can get the conclusions which are the same as the simulation data.

CONCLUSION

For the ill-posed inversion problem of PCS, considering the nonnegative of PSD, this study compared the nonnegative constraint features of Trust-Tikhonov and IPN-Tikhonov, combining with the respective advantages of two methods, Trust-IPN-Tikhonov is put forward in this study. This method inherits the advantage of Trust-Tikhonov and IPN-Tikhonov. Using above three methods, simulation data and experimental data were inverted. The inversion results demonstrate that inversion PSD of Trust-IPN-Tikhonov has higher accuracy and better smoothness, is more consistent with the true distribution.

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