J. Synchrotron Rad. (1999). **6**, 249–250

Linear inverse problem solution of the basic XAFS equation *via* the Wavelet–Galerkin method

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The basic XAFS equation given by Sayers, Stern and Lytle is solved by a linear inverse problem method instead of usual Fourier transform. The Wavelet-Galerkin discretization is utilized in our formulation for efficient calculations. As a result of model calculations, sufficiently exact solutions for radial distribution function are obtained with expansion by a small number of wavelets within the range of description of the basic XAFS equation.

Keywords: wavelet; inverse problem; XAFS equation.

1. Introduction

Since the proposition of the basic equation for the analysis of XAFS, or X-ray absorption fine structure by Sayers, Stern and Lytle (Sayers, Stern and Lytle, 1971; Lee and Pendry, 1975; Ashley and Doniach, 1975), structural informations are obtained mainly by the method of Fourier transform based on the assumptions that the approximation of phase shift by a linear function of wavenumber k and the utilization of a window function to limit the range of wavenumber k in the Fourier transform are justifiable. Even for some pure substances, the phase shift is an undulatory function of wavenumber k (Teo and Lee 1979) and Fourier transform is not adequate. As a typical example of usual XAFS analysis, the imaginary part of the windowed Fourier transform of Cu K-edge XAFS spectrum observed at Photon Factory in KEK is shown in figure 1. In the figure 1, there are corresponding peaks up to the third shell at approximately exact locations of each neighboring shell atoms. However, many pseudo peaks are also emerged and as an inherent characteristic of Fourier transform, the plot has impractical minus part. Moreover, proportions between the height of each peak do not reflect the correct number of atoms in each shell. In such cases, it is desirable to treat the basic XAFS equation from the point of view of the linear inverse problem to obtain more optimized solutions.

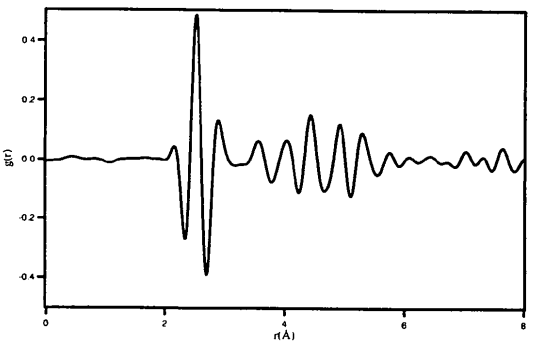

Figure 1
Windowed Fourier transform of Cu K-edge XAFS spectrum observed at Photon Factory in KEK.

Sayers Stern and Lytle formulated the basic XAFS equation by considering only single scattering in which the scattered electron is described by plane wave. In continuous form, for one kind of atom pairs, that equation is given as follows (Lee *et al.*, 1981),

$$\chi(k) = \frac{|f(k,\pi)|}{k} \int_0^\infty g(r) e^{-\frac{2r}{\lambda(k)}} \sin(2kr + \phi(k)) dr, \quad (1)$$

Here, $\chi(k), |f(k,\pi)|, \lambda(k)$ stand for the observed XAFS spectrum, the backscattering amplitude from neighboring atoms, and the electron mean free path, respectively. $\phi(k)$ is the total phase shift for the photoelectron. r and k are the distance from the center of excited atom and the wave number for the scattered electron. $g(r)$ is the unknown radial distribution function for neighboring atoms. This equation has the general form of the first kind Fredholm integral equation which can be generally solved with the method of linear inverse problems.

In the field of image restoration and computerized tomography, wavelet transform is incorporated into the method to solve linear inverse problems in each field for purposes of nonlinear noise filtering and efficient discretization of the problem. Wavelet transform is also acknowledged as an general alternative for windowed Fourier transform.

In the present article, a wavelet-Galerkin regularization algorithm to solve the single component basic XAFS equation is formulated and tested.

2. Methods

The k space part of the equation 1 is discretized using 256 fifth degree Daubechies wavelet(Daubechies 1992) bases which have the displacement and the scale parameter specified in the table 1.

Table 1
Ranges of wavelet displacement parameter for expansion of k space in each scale of wavelet subspaces. j_0 is the scale for the scaling function used in this expansion and is fixed to 1. j is the scale for the wavelet subspaces. This range is determined to cover the range of [1.5,15.5](Å^{-1}) in k space.

$j_0 = 1$	$j = 1$	$j = 2$	$j = 3$	Total number of wavelets
[-5,30]	[-1,34]	[2,65]	[8,127]	256

The r space part is discretized with the collocation method using 1024 sampling points between the range of [0.0, 8.0](Å). The discretized integral equation 1 takes the form of finite size matrix equation,

$$\chi = Kg. \tag{2}$$

The regularized solution of this matrix equation 2 is calculated by applying the Landweber-Friedman-Bialy iterative method (Landweber 1951),

$$g_{n+1} = g_n + \alpha K^*(\chi - Kg_n), \quad (\alpha > 0). \tag{3}$$

Here n is the number of iteration step, K^* is transposed K, g_n is the approximate solution for radial distribution function at nth step and α determines the rate of convergence in iteration. α is fixed to 5000.0 in our present calculation. As a stopping rule for iterations, we minimized the difference between the input spectrum and the reconstructed one,

$$\| Kg_n - \chi \| . \tag{4}$$

Zero vector is used as an initial guess for g_n in our calculations.

The condition on radial distribution function, $g(r) \geq 0$ is included by cropping the minus part of output $g_{n+1}(r)$ in each iteration, and then by using this cropped output as a guess for next step.

3. Test results

To estimate the basic performance of our scheme, a model copper K-edge XAFS spectrum is used as an input spectrum $\chi(k)$ for our test calculation. This model spectrum is produced by applying the equation 2 to the analytical model copper radial distribution function upto the seventh shell, which is defined as follows,

$$g(r) = C \sum_i A_i e^{-a(r-R_i)^2}. \tag{5}$$

The exponential parameter a and the coefficient C are fixed to 10.0 and 0.432529 respectively. The amplitude and the location parameters are given in the table 2.

Table 2
Amplitudes and locations of peaks for radial distribution function of copper.

Shell Number(i)	1	2	3	4
A_i	1.83679	0.459128	1.22459	0.459197
R_i(Å)	2.556	3.615	4.427	5.112
Shell Number(i)	5	6	7	
A_i	0.734816	0.204081	1.049451	
R_i(Å)	5.715	6.261	6.763	

The reconstructed radial distribution function for the model spectrum is shown in figure 2 with the original model distribution. The windowed Fourier transform of our model spectrum is also plotted in the same figure.

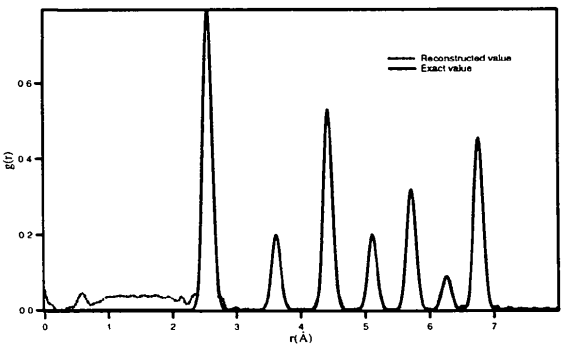

Figure 2
Reconstructed radial distribution function for model Cu K-edge XAFS spectrum. Exact oroginal model radial distribution function is also plotted for reference.

4. Conclusion

The basic XAFS equation is regularized by Wavelet-Galerkin method using Daubechies wavelet and solved by Landweber-Friedman-Bialy iteration method. The result of test calculation for our scheme is fairly improved in comparison with that of the Windowed Fourier Transform.

For practical use, our scheme is still insufficient because it is based on the too-much-simplified description of XAFS phenomena by the basic XAFS equation. It is to be modified to accomodate multiple scattering effects in future.

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(Received 10 August 1998; accepted 26 January 1999)