Atomic Image Reconstruction from Atomic Resolution Holography Using L_1 -Regularized Linear Regression*

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(Received 7 December 2015; Accepted 25 March 2016; Published 21 May 2016)

Inverse x-ray fluorescence holography provides new knowledge about the local atomic structure around dopants in crystals. The three-dimensional atomic image is reconstructed from the inverse x-ray fluorescence hologram using a calculation. The performance of the algorithm used for this calculation is important. The Barton method, which is based on the Fourier transform, is typically used. However, images reconstructed by this method sometimes contain artifacts. I developed an algorithm (SPEA-L1) using an iterative method with L_1 regularization and report its performance. [DOI: 10.1380/ejsnt.2016.158]

Keywords: Atom-solid interactions; Scattering; Computer simulations; X-ray scattering, diffraction, and reflection

I. INTRODUCTION

Local atomic structures have been intensively studied recently by inverse x-ray fluorescence holography (IXFH) [1–4] using synchrotron radiation facilities such as SPring-8. The method can measure the three-dimensional (3D) local atomic structure around dopants in crystals [5] and atomic position fluctuations [6, 7]. It provides new knowledge about the local atomic structure. The 3D atomic image is reconstructed from the hologram using a calculation. The performance of the algorithm used for the reconstruction calculation is quite important. The Barton method [8, 9], which is based on the Fourier transform, is typically used. However, images reconstructed by this method sometimes contain artifacts. I developed an algorithm using an iterative method with L_1 regularization and report its performance.

The recording process used in IXFH is described as follows. The sample is irradiated with x-rays. Part of the incident x-ray wave is scattered by atoms in the sample, and the scattered waves interfere with the incident wave. The holographic nanoscale interference pattern of the x-rays is formed in the sample. The emitted x-ray fluorescence is proportional to the photon density. The holographic pattern is measured as the intensity modulation of the fluorescence with respect to the incident angle. The hologram is given by

$$\chi(\mathbf{k}) \propto - \int \rho(\mathbf{r}) \frac{\cos(kr - \mathbf{k} \cdot \mathbf{r})}{r} d\mathbf{r}, \quad (1)$$

where $\chi(\mathbf{k})$ and $\rho(\mathbf{r})$ are the hologram function and electron density function, respectively. The origin of $\mathbf{r}$ is the atom emitting the x-ray fluorescence. Further, r and k are defined as $r = |\mathbf{r}|$ and $k = |\mathbf{k}|$.

The Barton method is typically used to reconstruct the 3D atomic image. The method is given by

$$u(\mathbf{r}) = -r \int \chi(\mathbf{k}) \cos(kr - \mathbf{k} \cdot \mathbf{r}) d\mathbf{k}, \quad (2)$$

where $u(\mathbf{r})$ is the reconstructed atomic image. This method is based on a type of Fourier transform.

In principle, to reconstruct the 3D atomic image, a 3D hologram is required because the amount of information is not changed by the Fourier transform. Therefore, an energy-tunable light source is required to measure the volume hologram by changing the wave number k of the incident x-rays. In addition, the basis function of Eq. (1) is not orthogonal. Consequently, artifacts sometimes appear in the reconstructed atomic image.

II. L_1 REGULARIZATION

I developed an algorithm, SPEA-MEM [10], to reconstruct a 3D atomic image from a photoelectron hologram. SPEA-MEM is a fitting-based algorithm using the maximum entropy method. The maximum entropy is a good way to solve this problem. However, in this paper I investigate another method of checking the performance. The developed algorithm uses L_1 -regularized linear regression [11], which can lead to sparse solutions. The algorithm is called SPEA-L1 (scattering pattern extraction algorithm with L_1 regularization).

When the atomic scattering factor $f(\theta)$ is introduced, the hologram is given by [1]

$$\chi(\mathbf{k}) = - \sum_i \frac{f(\theta_{\mathbf{a}_i \mathbf{k}}) \cos(ka_i - \mathbf{k} \cdot \mathbf{a}_i)}{a_i}, \quad (3)$$

where $\mathbf{a}_i$ represents the i -th atom's position. $\theta_{\mathbf{a}_i \mathbf{k}}$ is the angle between $\mathbf{a}_i$ and $\mathbf{k}$. This equation can be extended by introducing the atomic distribution function $g(\mathbf{r})$ as

$$\chi(\mathbf{k}) = - \int g(\mathbf{r}) f(\theta_{\mathbf{r} \mathbf{k}}) \cos(kr - \mathbf{k} \cdot \mathbf{r}) d\mathbf{r}. \quad (4)$$

When the atomic distribution function is set to $g(\mathbf{r}) = \sum_i \delta(\mathbf{r} - \mathbf{a}_i)/r$, Eq. 4 gives the hologram. The atomic distribution function $g(\mathbf{r})$ is sparse. Therefore, L_1 -regularized linear regression is applicable. When $g(\mathbf{r})$ is described using voxels, the equation is modified as

$$\chi(\mathbf{k}_j) = - \sum_i g(\mathbf{r}_i) f(\theta_{\mathbf{r}_i \mathbf{k}_j}) \cos(k_j r_i - \mathbf{k}_j \cdot \mathbf{r}_i). \quad (5)$$

* This paper was presented at the 10th International Symposium on Atomic Level Characterizations for New Materials and Devices '15, Shimane Prefectural Convention Center (Kunibiki Messe), Matsue, Japan, October 25-30, 2015.

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FIG. 1. Simulated x-ray fluorescence hologram of 2 nm Fe cluster. Photon energy is 7500 eV.

To obtain $g(\mathbf{r}_i)$, L_1 -regularized linear regression is applied here. Its evaluation function is given by

$$E = \frac{1}{2}E_1 + \frac{\lambda}{2} \sum_i |g(\mathbf{r}_i)|, \quad (6)$$

$$E_1 = \sum_j |\chi(\mathbf{k}_j) - \hat{\chi}(\mathbf{k}_j)|^2, \quad (7)$$

where $\hat{\chi}(\mathbf{k})$ is defined as the observed hologram.

The parameter λ is controls the sparseness of $g(\mathbf{r})$. To obtain the atomic image, a calculation to minimize the evaluation function E is performed. The iterative calculation is given by

$$g^{(n+1)}(\mathbf{r}) = g^{(n)}(\mathbf{r}) - \alpha \frac{\partial E^{(n)}}{\partial \mathbf{r}}, \quad (8)$$

where n is the index for iteration. The parameter α is optimized using the gradient method. In addition, a non-negative boundary condition is applied to the voxel value $g(\mathbf{r}_i)$.

III. RESULTS AND DISCUSSION

To test the algorithms, 10 simulated holograms of a 2 nm Fe cluster at photon energies of 7500–1200 eV were prepared. Figure 1 shows a simulated hologram of an iron crystal. The energy of the incident x-rays is 7500 eV. The used cluster size is quite smaller than the x-ray mean free path in solid. According to Eq. (1), the hologram caused by distant atom from the emitter has weak amplitude and high frequency. Therefore, the effect of the distant atom quite rapidly decreased in comparison with the mean free path. It is better to use the 3 nm–5 nm cluster to reproduce the experimental XFH. Since the aim of this paper is to show the performance of SPEA-L1, the holograms that cause the artifacts in the use of the Barton method were prepared.

The 3D atomic image reconstructed using the Barton method is shown in Fig. 2. One can reconstruct any place of the atomic image because the each volume $u(\mathbf{r}_i)$ can be independently calculated by using Eq. (2). The reconstructed volume is set to be $\pm 0.5 \text{ nm}^3$ to see the structure around the emitter.

FIG. 2. Atomic image of Fe crystal reconstructed using Barton method.

The Fe crystal structure is reconstructed. However, there are many artifacts; for example, a large artifact appears between the first and fourth nearest-neighbor (NB) atoms. In addition, the atomic image sizes, which correspond to the intensity of the atomic image, are different. The atomic image sizes of the first and fourth NBs are large, and that of the third NB is small. This phenomenon is caused by the non-orthogonal basis function in Eq. (1).

Then, SPEA-L1 is tested. Here, the atomic image with $\pm 0.5 \text{ nm}^3$ region, which is smaller than the cluster size is utilized in order to check the artifact caused by the atoms located outer side of the atomic image.

The estimation of the parameter λ is important. When λ is increased, the amount of zero in voxel is increased. In this calculation, the parameter λ was set using

$$\lambda = \beta \max\left(-\frac{\partial E_1^{(n)}}{\partial \mathbf{r}_i}\right), \quad (9)$$

and the parameter β was set to 0.9. In this case, most values of $-\partial E/\partial \mathbf{r}$ become negative.

The reconstructed atomic images for the N -th iteration

FIG. 3. Atomic images reconstructed using SPEA-L1. RMSE is displayed at the top.

and the root mean squared error (RMSE) ($= \sqrt{E_1}$) are shown in Fig. 3.

As the iterations progress, the atomic image is repro-

duced from the inside. After five iterations ($N = 5$), the atoms from the first to fifth NBs are clearly reconstructed. The atomic image sizes of the first to fifth atoms are almost the same. There are no large artifacts. Some small artifacts appear near the first and third NB atoms. After further iterations are performed, the number of artifacts increases. These artifacts appear for two reasons. The first reason is a type of sidelobe effect of the Fourier transform. The SPEA-L1 algorithm is a fitting-based algorithm; however, the effects of the window function of the Fourier transform appear. In Eq. (2), the integral region is finite, and the window function is not introduced. In this situation, the sidelobe is enhanced. The artifacts in the image at $N = 5$ may be caused by this sidelobe effect. The second reason is the so-called overfitting effect, which sometimes appears in fitting-based algorithms. This algorithm might search to reproduce the hologram caused by the atoms located outer side of the image. To suppress overfitting, the parameter λ has been introduced. The method of calculating λ is important. In this paper, a simple method of obtaining λ was used. In the early stage of the iteration, this method works well. However, it is not suitable after further iterations. A better way is required to obtain better atomic images.

IV. CONCLUSION

The 3D atomic image reconstruction algorithm SPEA-L1, which is a fitting-based algorithm using L_1 regularization, is developed. In this paper, the parameter λ is obtained using Eq. (9). In this way, a very clear atomic image is obtained after 5 – 7 iterations. This image still involves small artifacts caused by a type of sidelobe effect of the Fourier transform. When further iterations are performed, the number of artifacts increases because of the overfitting effect. SPEA-L1 should be further developed by introducing a window function and a better method of setting the parameter λ .

V. ACKNOWLEDGEMENT

The author is deeply grateful to Prof. Koichi Hayashi, Prof. Shinya Hosokawa, and Prof. Naohisa Happo for helpful discussions about the theoretical and experimental development of IXFH. This work was supported by JSPS Grant-in-Aid for Scientific Research on Innovative Areas “3D Active-Site Science,” Grant Number 26105013 2604.

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