Application of the Allen – Oxley iterative method of phase retrieval to series of images obtained by focus variation

Iván Lobato[1], Eduardo Montoya[2]

[1] Facultad de Ciencias, Universidad Nacional de Ingeniería, Av. Tupac Amaru 210, Lima 25, Perú
[2] Departamento de Química, Instituto Peruano de Energía Nuclear, Av. Canadá 1470, Lima 41, Perú

Resumen

Se describe la aplicación del método iterativo de recuperación de fase a una serie de imágenes obtenidas a diferentes valores de desenfoque. Se muestra que el esquema iterativo es robusto en presencia de discontinuidades en la fase. La aplicación de este método a un conjunto de imágenes experimentales tomadas usando rayos-X es investigada. Se discute brevemente el programa de computadora que puede reproducir la simulación y el análisis experimental de los datos de las imágenes.

Abstract

The application of the iterative method of phase retrieval to series of images obtained at different defocus values is described and discussed. It is shown that the iterative scheme is robust in the presence of discontinuities in the phase. The application of these methods to a set of experimental images taken using X-ray imaging is investigated. A computer program which can reproduce the simulations and analyse experimental image data is briefly discussed.

1. Introduction

Non-interferometric determination of the phase of quantum mechanical and classical wave fields (i.e. phase retrieval) is a topic of current interest in a number of areas where either phase imaging or structure retrieval is an issue. For example, phase measurement is topical for optical, X-ray, neutron, electron and atom wave fields. However, these measurements are made possible by the fact that the thin Transmission Electron Microscopy (TEM) sample acts as a phase object on the electron wave and that the phase shift is directly related to the sample inner potential. In this paper we develop and apply iterative method of phase retrieval from series of images taken at different defocus values.

The accurate determination of size distributions on materials consisting in metal nanoparticles which are embedded in a matrix is not always straightforward, from TEM or HRTEM images. In a paper by Donnadieu *et al.* [1] an approximated method of phase retrieval, based on an approximated solution of the transport intensity equation (TIE), has been proposed for the imaging of dense nanodot assemblies. According to the cited authors, the method is of easy implementation and can be used in standard transmission electron microscopes. Besides the determination of the heights and chemical composition of the nanodots, discussed in [1], the knowledge of the phase of a HRTEM image would allow a better determination of the size and shape of the particles. The price of the approximations made in the cited reference is a low resolution which is determined by the defocus between images. On the other hand the minimal useful defocus depends on the changes in the image while defocusing, which is depending on the studied material. In practical terms, a resolution of 2.5 nm has been reported in reference [1] for a difficult material comprised by 10 nm silicon nanodots deposited onto an oxidized Si(100) surface. The low resolution would impair the good determination of the size and shape of very small particles like those mentioned before.

The present work describes the implementation of the iterative method proposed by Allen and Oxley [2], and its application to the case of the afore mentioned silicon nanodots deposited onto an oxidized Si(100) surface, using the TEM images published by Donnadieu *et al.* [1].

* Corresponding author: emontoya@ipen.gob.pe

2. Iterative Method of Phase Retrieval

Following reference [2], the starting point is the Schrödinger equation for the propagation of a wave in free space and in three dimensions,

$$\left(\nabla^2 + k^2\right)\Psi(\vec{r}) = 0 \qquad (1)$$

Where *k* is the wave number and is related to the wavelength of the radiation by k = 2π/λ. Let us assume that the wave function $\psi(\vec{r})$ can be considered as a perturbation of a plane wave traveling along the z direction and can be written in the form

$$\psi(\vec{r}_\perp, z) = \exp(ikz)\xi(\vec{r}_\perp, z) \qquad (2)$$

Where $\vec{r}_\perp$ is a vector in the x-y plane and perpendicular to the z direction. Then with the paraxial approximation that the second partial derivative of $\xi(\vec{r}_\perp, z)$ with respect to z is small, i.e. $\partial^2(\vec{r}_\perp, z) \approx 0$, the result is

$$\left(\nabla_\perp^2 + 2ik\partial_z\right)\xi(\vec{r}_\perp, z) = 0 \qquad (3)$$

Where $\nabla_\perp$ operates in the x-y plane y ∂_z should be read as a short hand notation for $(\partial/\partial z)$. From the equation (3) we have that the formal solution is:

$$\nabla_\perp^2\ \xi(\vec{r}_\perp, z) = -2ik\,\partial_z\,\xi(\vec{r}_\perp, z)$$

$$\frac{i\,dz}{2k}\nabla_\perp^2\ \xi(\vec{r}_\perp, z) = d\,\xi(\vec{r}_\perp, z) \qquad (4)$$

$$\xi(\vec{r}_\perp, z) = \exp\left(\frac{i\,\Delta z\,\lambda}{4\pi}\nabla_\perp^2\right)\xi(\vec{r}_\perp, z=0)$$

From which, using the properties about derivatives of Fourier transforms, it is shown that

$$\xi(\vec{r}_\perp, z=\Delta z) = F^{-1}\left[\exp\left(-i\,\Delta z\,\lambda q_\perp^2\right) F\left[\xi(\vec{r}_\perp, z=0)\right]\right] \qquad (5)$$

The last equation describes the free space propagation of the wave function from the plane at z = 0 to that at z = Δz. The wave functions can be written in terms of intensity and phase as,

$$\xi(\vec{r}_\perp, z) = I^{1/2}(\vec{r}_\perp, z)\exp\left[i\phi_\perp(\vec{r}_\perp, z)\right] \qquad (6)$$

In the iterative method of Allen and Oxley, the equations (5) and (6) are used in the following way: Starting at the principal (central) plane, a wave function is constructed, via equation (6), from the intensity of the known image and a guess for the phase map (in practice zero everywhere). This wave function is propagated to the next plane using the equation (5). Unless the initial guess for the phase is correct, the intensity of the propagated function will not agree with that of the known image at this plane. So the intensity of the propagated wave function is replaced with the known values. The step of propagation – intensity correction is repeated until reaching the last plane. Then the steps are applied in back propagation, crossing the central plane and continuing until the first plane is reached and finally the steps are applied from the first plane until the central plane is reached again. All this constitutes one cycle of the iterative procedure and after completed the sum squared error (SSE) is calculated according reference [3], using the equation (7). Then the iterations continue until either a convergence criterion, defined in terms of SSE, is satisfied or the SSE not longer decreases and stagnation occurs. Following reference [2] the Allen – Oxley iterative algorithm is represented as a schematic diagram in Figure 1, for a series of five images.

$$SSE = \frac{\sum_{i=1}^{N}\sum_{j=1}^{N}\left(\sqrt{I_{found}(x_i, y_j)} - \sqrt{I_{known}(x_i, y_j)}\right)^2}{\sum_{i=1}^{N}\sum_{j=1}^{N} I_{known}(x_i, y_j)} \qquad (7)$$

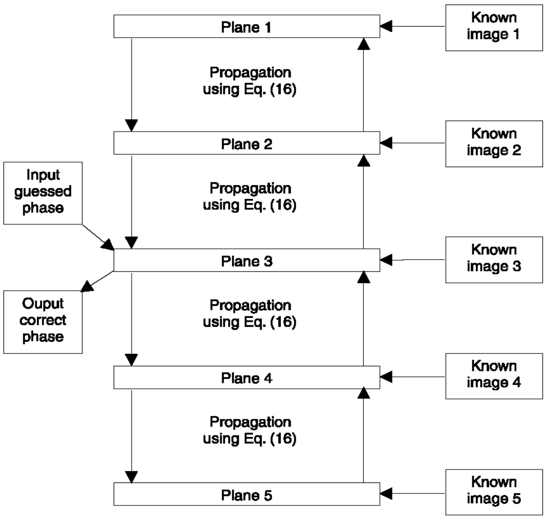

Figure 1: Schematic diagram of the Allen-Oxley iterative method, taken from reference [2].

To test the performance of the Allen – Oxley iterative method, the authors of reference [2] used the following model image and phase maps (assumed to be at zero defocus). The image is given by

$$I(x,y) = 1.0 - 0.9\Big(\exp\big\{-b^2\big[(x-x_1)^2 + (y-y_1)^2\big]\big\} + \exp\big\{-b^2\big[(x-x_2)^2 + (y-y_2)^2\big]\big\}\Big) \quad (8)$$

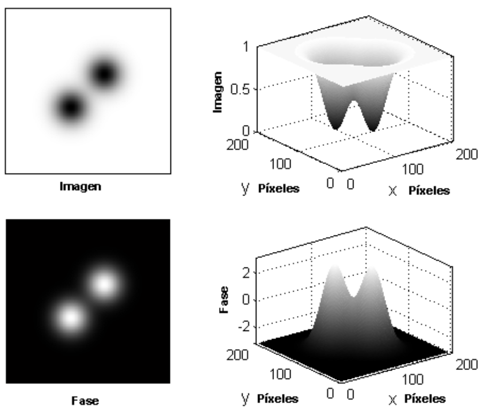

Figure 2: Input model image and phase given by Eqs. (8) and (9) (assumed to be at zero defocus) and shown as both grey scale and surface plots.

where b = 0.04. The location of the centre of the first Gaussian peak is at (x_1, y_1) with $x_1 = 2n_x/5$ and $y_1 = 3n_y/5$ and n_x and n_y are the numbers of pixels along the x and y directions respectively. The second Gaussian is centred at (x_2, y_2) with $x_2 = 3n_x/5$ and $y_2 = 2n_y/5$. The phase almost covers the range between -π and π and is given by

$$\phi(x,y) = 0.95\left\{\frac{2\pi}{0.9}\left[1.0 - I(x,y)\right] - \pi\right\} \quad (9)$$

The factor 0.95 is used by the authors of the cited reference to avoid phase wrapping due numerical errors during calculations.

3. Results and Discussion

The two first columns of the Figure 3 shows a series of simulated images and its corresponding phases, prepared following the reference [2], using the equations (8) and (9) followed by the application of the equations (6) and (5). The obtained images and phases reproduce very well those shown in two first columns of the Figure 4 of the reference [2]. Following reference [2], for each intensity image and its corresponding phase, a defocus series of 5 intensity images was "prepared" by calculation, using a defocus step of 10 mm, keeping the initial image at the central plane.

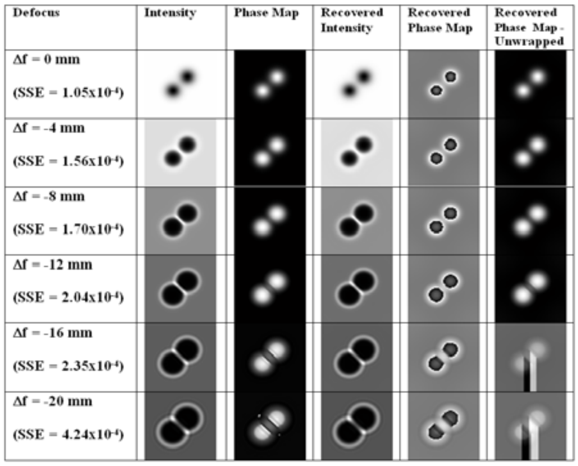

Figure 3: Defocus series and phase retrieval, by using the Allen – Oxley iterative algorithm implemented in the present work. The SSE values for the recovered intensity image, after N=1000 iterations, are given inside parentheses. The intensity and Phase maps prepared by free space propagation. The recovered phase maps have resulted accurate in all cases.

The images have a size of 256 x 256 pixels and represent an object of 1 mm by 1 mm, imaged with a HeNe laser (λ = 632.8 n). Two differences between the images prepared in the reference [2] and the present ones are the size, which is 193x193 pixels in reference [2], and the value of the parameter *b* in the equation (8) which is b = 0.04 in the present work, being b = 0.0028 in the cited reference. In the present work, the changes in size were made for a better performance of the Fast Fourier Transform operations and the changes in b were done for keeping the aspect of the prepared images as close as possible to the aspect of the images shown in the Figure 4 of the cited reference.

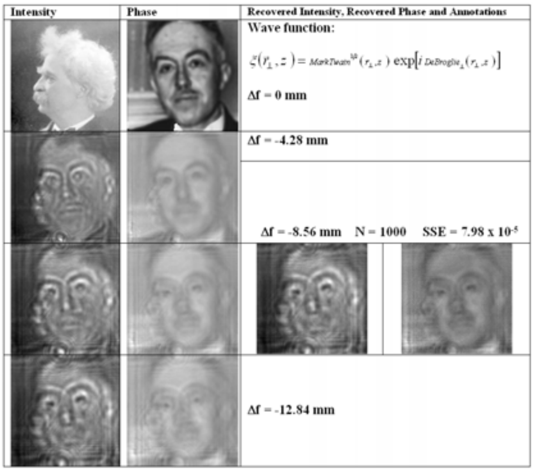

Figure 4: Defocus series prepared by free space propagation, after reference [5], and recovered intensity image and phase map, by using the Allen – Oxley iterative algorithm implemented in the present work.

The Figure 4 shows the intensity images and corresponding phase maps, obtained by free space propagation, following the reference [5]. The pictures of Mark Twain (used as intensity image) and L. de Broglie (used as phase map), in the Figure 1 of the cited reference were used as the starting image and phase map at Δf = 0 mm. The images have a size of 256x256 pixels (1.28 mm by 1.28 mm), and radiation of λ = 632.8 nm has been used. Again the obtained images and phases reproduce very well those shown in the Figure 3 of the reference [5].

The last column of Figure 3 shows the retrieved phase, obtained by application of the implemented Allen – Oxley iterative method to the images of the first column. The upper part of Figure 5 shows the retrieved intensity and phase corresponding to the three last images of Figure 4, where the original image of Mark Twain had been excluded and the image at Δf = -8.56 mm was used as the central image.

The lower part of the figure shows the intensity and phase obtained by back propagation to the plane at Δf = 0 mm. The recovered phase for the series of images from Figure 5 is shown in the right part of the figure. The value of the SSE after N = 1000 iterations is shown for each case.

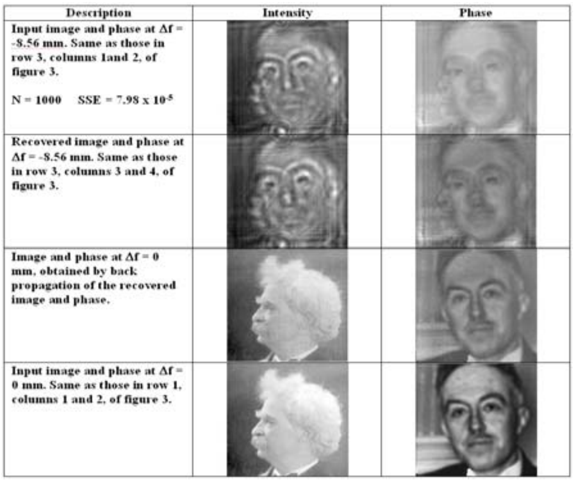

Figure 5: Recovered – back propagated intensity and phase, by using the Allen – Oxley iterative algorithm implemented in the present work, after reference [5]. Some pictures in figure 4 have been repeated for easier comparison.

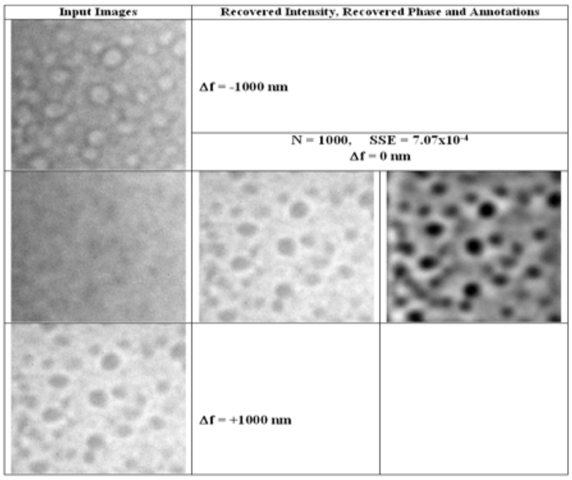

Figure 6: Recovered intensity image and phase map, by using the Allen – Oxley iterative algorithm, starting from the retrieved versions of the images from the figure 3 of the reference [1], in PDF version. The retrieved images have a size of 75.68 nm by 75.68 nm (256 pixels by 256 pixels) and correspond to a specimen of silicon nanodots deposited onto an oxidized Si(100) surface.

The left part of Figure 6 shows retrieved versions of the images from the Figure 4 of the reference [1], in PDF version. The images have a size of 75.68 nm by 75.68 nm (256 pixels by 256 pixels) and correspond to a specimen of silicon nanodots deposited onto an oxidized Si(100) surface. A microscope operating at 300 kV was used by the authors of the reference [1] and the defocus values are Δf = -1000 nm, Δf = 0 nm and Δf = 1000 nm. The recovered intensity image and phase map are shown in the right part. The value of the SSE after N = 1000 iterations is shown for each case.

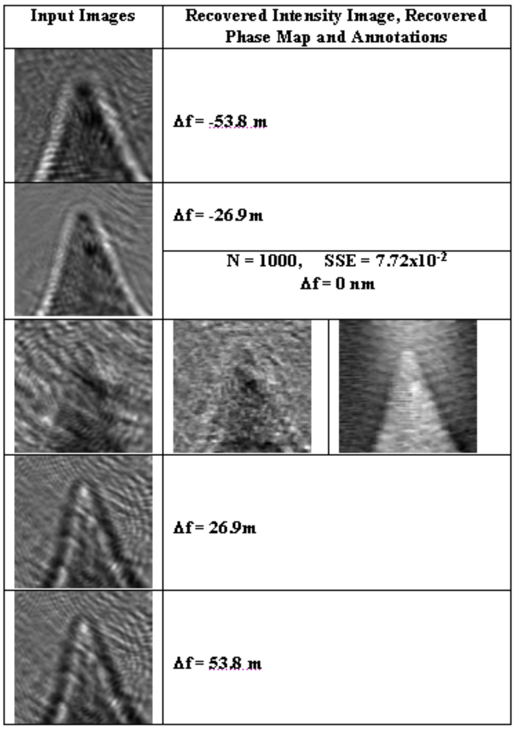

Figure 7: Recovered intensity image and phase map, by using the Allen – Oxley iterative algorithm implemented in the present work, starting from the retrieved versions of the images from the figure 3 of the reference [2], in PDF version. Phase retrieval from a through focal series of X-ray images of a silicon tip from an AFM. The X-rays had an energy of 1.83 keV (1 = 6.77 A). Each image, originally 70 x 70 pixels, representing a linear size of 1.69 mm.

The left part of Figure 7 shows retrieved versions of the images from the Figure 4 of the reference [2], in PDF version. The images have a size of 1.69 mm by 1.69 mm (100 pixels by 100 pixels) and correspond of X-ray images of a silicon tip from an AFM. The X-rays had an energy of 1.83 keV (1 = 6.77 A). Each image, originally 70 x 70 pixels, representing a linear size of 1.69 mm and the defocus values are Δf = -53.8m, Δf = -26.9 m, Δf = 0 m, Δf = 26.9m and Δf = 53.8m. The recovered intensity image and phase map are shown in the right part. The value of the SSE after N = 1000 iterations is shown for each case.

The results shown in the first two columns of the Figure 3 as well as in the Figs. 4 and 5 are evidence that the implemented free space propagator is working properly. Also the results of phase retrieval shown in the Figs. 5, 6 and 7 are evidence that the present implemented version of the Allen – Oxley iterative method is working properly. This can be appreciated specially in the case of the Figure 4, in accordance with what has been stated in reference [5] about that the iterative method is robust in presence of first order vortices. The results of phase retrieval shown in Figure 3 have some differences with the results shown in the last column of the Figure 4 in the reference [2]. In first place, a strong wrapping of the phase is observed and in second place, the results in presence of vortices are not as good as the results presented in reference [2]. Also artifacts due the presence of first order vortices can be appreciated in the phase maps shown in the last column of Figure 3. As stated in the reference [2] the phase unwrapping in presence of vortices is not straightforward. It has to be pointed out that, when using images of 193x193 pixels, no differences with the results described in reference [2] are observed.

4. Conclusions

We have implemented and applicated the Allen – Oxley iterative method of phase retrieval with good results in all the tested cases. We have illustrated phase retrieval on defocused images for the specific example of nanodot assemblies and the phase retrieval from a through focal series of X-ray images of a silicon tip from an AFM. The step next is to apply the Iterative Method of Phase Retrieval to a imagine set of low resolution that it will be obtained in the transmission electron microscopy (TEM) of the IPEN.

5. References

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