Complex Electron Wave Reconstruction Using Parameter Estimation

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***Abstract*— A new method is proposed for the reconstruction of the complex valued exit wave of a periodic specimen in a transmission electron microscope. The method uses a series of images recorded at different defoci. From these, inherently noisy, images the parameters defining the wave are estimated. The method keeps the number of parameters as small as possible. In addition, in simulations, it has been found to always produce the exit wave estimate fitting best to the images recorded.**

***Index Terms*— Electron microscopy, Fourier techniques, identification, image reconstruction, maximum likelihood, parameter estimation.**

I. Introduction

The purpose of this paper is to present a new method for the reconstruction of the real and imaginary part of the complex exit wave of the specimen in a transmission electron microscope (TEM). The wave is used for the assessment of the structure of the material of the specimen. The TEM image is different from the complex exit wave for two reasons. In the first place, the magnifying system of the TEM transfers the exit wave to the camera level. This transfer is modeled by a linear transfer function which is supposed to be known for known microscope settings. Then, at the camera level, the transferred complex wave is transformed into an image, that is, into intensities. This is supposed to be a modulus square operation. The image pixels thus produced are the observations. They are modeled as stochastic variables since they are finite time electron counts. Other statistical contributions to the observations, like instrumental noise, are negligible in modern TEM's [1].

The existing reconstruction methods reconstruct the exit wave from a series of at least two images, measured at different, known defoci [2]–[5]. The purpose of the defocus series is to use the known dependence of the transfer function upon the defocus to ensure that all frequencies within the available bandwidth are more or less equally taken into account [6].

A common characteristic of the existing methods is that they reconstruct the exit wave in every point of the measured images. Therefore, every point of the wave is a parameter to be estimated. This number of parameters is drastically reduced in the method presented in this paper. Assuming that the specimen structure is periodic, as is done with respect to almost every specimen described in the literature, the method estimates the complex Fourier coefficients of the exit wave, which in that case is periodic as well. The estimated complex Fourier coefficients define fully the exit wave.

The advantage of the proposed method is that the number of Fourier coefficients, that is, the number of parameters is fixed. This number does not increase with the number of pixels measured. All observations are used to reconstruct the wave on a single rectangle with sides equal to the periods in the corresponding coordinate directions. This rectangle is the projected unit cell of the specimen material. The only effect of using more pixels is improvement of the precision of the reconstructed exit wave. Furthermore, as opposed to the existing methods, the present method benefits from the fact that in practical electron microscopy the number of Fourier coefficients characterizing the projected unit cell is known to be very small.

The organization of this paper is as follows. In Section II, the intensity observations are modeled and the problem is stated. In Section III, a maximum likelihood estimator for the Fourier coefficients of the exit wave is derived. Aspects of numerical implementation of this estimator are the subject of Section IV. Results from simulated statistical intensity observations are described in Section V. A discussion and conclusions are presented in Section VI.

II. Modelling the Observations

In transmission electron microscopy, the complex electron wave present at the exit interface of the irradiated specimen is called the exit wave. In this paper, it will be assumed that the specimen has a rectangularly periodic crystalline structure. Then the exit wave is rectangularly periodic as well. Therefore, it is described by the Fourier series

$$f(x,y) = \sum_{k,\ell} \gamma_{k\ell} \exp j(\omega_k x + \omega_\ell y).$$

In this expression, $f(x,y)$ is the complex exit wave, x and y are the spatial coordinates, j is equal to $\sqrt{-1}$ and ω_k and ω_ℓ are the spatial radian frequencies in the x and y direction, respectively, defined as $\omega_k = 2\pi k/X$ and $\omega_\ell = 2\pi \ell/Y$, where k and ℓ are the harmonic numbers of the harmonics present, X and Y are the periods, and the $\gamma_{k\ell}$ are the complex Fourier coefficients.

Subsequently, the wave is magnified and transferred to camera level. This operation is described using a linear transfer function $H(\omega_x, \omega_y)$ where ω_x and ω_y are radian spatial frequencies in the x and y direction, respectively. For brevity, the notation $H(k,\ell)$ will be used for $H(\omega_k, \omega_\ell)$. Then the

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transferred electron wave $g(x,y)$ is described by

$$g(x,y) = \sum_{k,\ell} H(k,\ell)\gamma_{k\ell} \exp j(\omega_k x + \omega_\ell y).$$

Finally, at camera level, the transferred wave is transformed into intensities by a modulus square operation

$$i(x,y) = |g(x,y)|^2.$$

It will be supposed that the intensities are measured at the known locations (x_p, y_q), $p = 1,\ldots,P$ and $q = 1,\ldots,Q$. Like the existing reconstruction methods, the proposed method uses a defocus series of images. This is a series of M images made at different, known defoci. The transfer function depends on the defocus in a known way. The notation $H_m(k,\ell)$ will be used for $H(k,\ell)$ at the mth defocus. Similarly, quantities like $i(p,q)$ and $g(p,q)$ at the mth defocus will be denoted as $i_m(p,q)$ and $g_m(p,q)$ where p and q are abbreviations of x_p and y_q.

Next suppose that at the mth defocus the intensity observations $w_m(p,q)$, $p = 1,\ldots,P$, and $q = 1,\ldots,Q$ have been made. Then these observations are modeled as a realization of stochastic variables with expectation

$$\begin{aligned} E[w_m(p,q)] &= i_m(p,q) \\ &= \left| \sum_{k,\ell} H_m(k,\ell)\gamma_{k\ell} \exp j(\omega_k x_p + \omega_\ell y_q) \right|^2. \end{aligned} \tag{1}$$

Therefore, the $w_m(p,q)$ are equal to the sum of this expectation and a zero-mean error. The expression also shows that the observations are quadratic in the unknown Fourier coefficients of the exit wave. These Fourier coefficients are taken as the unknown parameters to be estimated from the observations at the various defoci. Once they have been estimated, an estimate of the exit wave $f(x,y)$ can be made by Fourier synthesis.

Thus the measurement of the exit wave has been reformulated as estimation of the complex parameters $\gamma_{k\ell}$ from a series of images $w_m(p,q)$, $p = 1,\ldots,P$, and $q = 1,\ldots,Q$ with $m = 1,\ldots,M$. A solution of this estimation problem is proposed in the next section.

III. Using the Model of the Observations for Exit Wave Estimation

If the probability density function of the observations $w_m(p,q)$ is known, the maximum likelihood estimator for the $\gamma_{k\ell}$ may be chosen. This estimator is attractive since it is easy to construct directly from the probability density function of the observations and is, under general conditions, asymptotically most precise [7].

The maximum likelihood estimator is defined as the estimator maximizing the so-called (log) likelihood function of the unknown parameters given the observations. As an illustration, suppose that the intensity observations $w_m(p,q)$ are independent and have a Poisson distribution [8]. These assumptions are often made and are based on the fact that the intensity observations are largely independent electron counts. Under these assumptions, it is easily shown that the likelihood function of the unknown Fourier coefficients given the observations $w_m(p,q)$ for all m, p, and q is described by

$$\begin{aligned} L(c) = \sum_m \sum_{p,q} &-i_m(p,q) \\ &+ w_m(p,q)\ln(i_m(p,q)) - \ln(w_m(p,q)!) \end{aligned} \tag{2}$$

with

$$i_m(p,q) = \left| \sum_{k,\ell} H_m(k,\ell) c_{k\ell} \exp j(\omega_k x_p + \omega_\ell y_q) \right|^2. \tag{3}$$

To emphasize that the Fourier coefficients have now become variables instead of exact parameters, all $\gamma_{k\ell}$ have been replaced by corresponding $c_{k\ell}$. The vector c is the vector of all $c_{k\ell}$. The equations show that the likelihood function is a nonquadratic, nonlinear function of the $c_{k\ell}$. Therefore, maximizing it requires an iterative numerical method. The equations also show that the nonlinear dependence of $L(c)$ on c is complicated. As a result, it is difficult to analyze. However, numerical experiments strongly suggest the presence of relative maxima. For the usual fast, local numerical optimization methods this means that the absolute maximum, that is the maximum likelihood estimate, is found only if accurate initial values for the parameters are available. The generation of such initial values is described in the next section.

IV. Initial Values for the Parameters

For the generation of the initial values for the parameters, the following procedure is proposed. In a first step, the least-squares criterion

$$\sum_m \sum_{p,q} \{w_m(p,q) - i_m(p,q)\}^2 \tag{4}$$

with $i_m(p,q)$ described by (3) is minimized with respect to c. In a second step, the least-squares estimate thus found is used as initial condition for maximizing the likelihood function (2).

The advantages of this approach are the following. The intensity model (3) is quadratic in the Fourier coefficients $c_{k\ell}$. Therefore, the least-squares criterion is a real, multidimensional, quartic polynomial in the real and imaginary parts of the $c_{k\ell}$. Then the intersection of the criterion with any vertical two-dimensional plane has either two minima and a maximum in between or a single minimum. In view of the symmetry properties of the quartic multidimensional polynomial forming the least-squares criterion, this implies that this criterion might have only one distinct, and therefore absolute, minimum. Anyway, this is what has been found in all simulations with two or more defoci. For a particular set of observations, the least-squares procedure always converged to the same minimum, irrespective of the initial conditions. This happened both for simulated exact observations and Poisson distributed ones.

Although the initial conditions do not affect the solution, they influence the number of iterations required for convergence and, therefore, the computation time. If no *a priori* knowledge about the $c_{k\ell}$ is available, initial conditions of more or less the right order of magnitude are generated as

TABLE I
HARMONIC NUMBERS AND FOURIER COEFFICIENTS IN EXPERIMENT #1

-2	-1	0	1	2
- 0.75 - 0.5j	1.5 + 2j	1	1 + j	0.5 - j

follows. First, it is observed that the average value i of the exact $i_m(p,q)$ over all p, q, and m is equal to

$$\frac{1}{M}\sum_{k,\ell}|\gamma_{k\ell}|^2\sum_m|H_m(k,\ell)|^2.$$

In this expression, the $H_m(k,\ell)$ are known. If all $\gamma_{k\ell}$ would have the same absolute value $|\gamma|$, this value would have to satisfy

$$|\gamma|^2 = i/H \quad \text{with} \quad H = \frac{1}{M}\sum_{k,\ell}\sum_m|H_m(k,\ell)|^2. \qquad (5)$$

Substituting the average of all observations $w_m(p,q)$ for the average i of all exact observations $i_m(p,q)$ in (5), produces an approximation $|c|^2$ to $|\gamma|^2$. Next the initial conditions for the $c_{k\ell}$ are generated as $|c|\exp(j\phi_{k\ell})$ where $\phi_{k\ell}$ is a random number uniformly distributed on $(0,2\pi)$. Thus it is achieved that the initial $c_{k\ell}$ are in the mean square sense of the right order of magnitude.

In conclusion, a procedure has been devised for the generation of initial values for the minimization of the least-squares criterion (4). The results of this minimization may subsequently be used as initial values for the minimization of the likelihood function, for example, that defined by (2) and (3).

V. SIMULATION RESULTS

A. Simulation Experiment #1

This experiment is a first investigation of the properties of the proposed estimator. The images are one-dimensional to reduce the computational effort and to simplify the display. The number of images in a series (M) is chosen equal to two. The number of pixels in an image is equal to 50. The observations are Poisson distributed. The harmonics present in the exit wave and the corresponding complex Fourier coefficients are given in the first and second row of Table I, respectively. Changing the phase angles of all Fourier coefficients by the same amount does not change the corresponding intensities. Therefore, to make the parameterization unique, the phase angle of one of the Fourier coefficients has to be fixed. This is done by assuming that the constant term of the Fourier series is real. Thus there are four complex parameters and a real one to be estimated, that is, nine real parameters in all. Notice that the complex Fourier coefficients for opposite harmonic numbers are not conjugate since the wave is not real.

The transfer functions are chosen as follows. The modulus is equal to one in all cases. The phase angles are generated by a random number generator and are uniformly distributed on $(0,2\pi)$. The number of periods observed is equal to 2.2 and, since the number of pixels is equal to 50, the number of pixels per period is equal to 22.7. The reason why noninteger numbers have been chosen is that this makes the simulation more realistic. In this experiment, the required numerical optimization procedures are carried out using the conjugate gradient method [9].

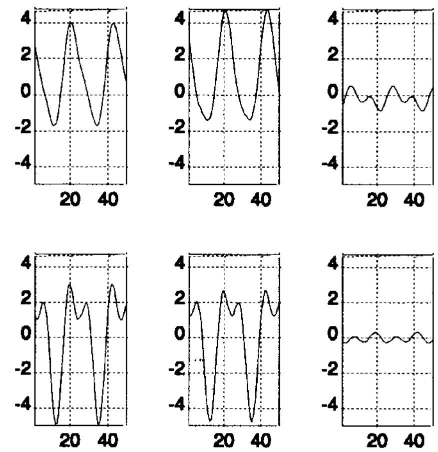

Fig. 1. Simulation experiment #1. Real and imaginary parts of exact and reconstructed exit wave, and reconstruction error.

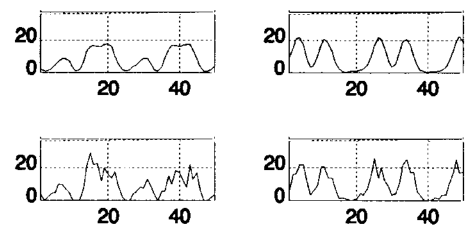

Fig. 2. Simulation experiment #1. Exact (top) and Poisson distributed (bottom) intensities.

From the exact Fourier coefficients of Table I, the exact complex exit wave is computed. The top and bottom figures in the first column of Fig. 1 show its real and imaginary part, respectively. The exact Fourier coefficients and the generated values of the transfer functions are subsequently substituted in (1) to compute both exact images of the defocus series. The results are shown in the top row of Fig. 2. The proposed method and the software were tested by first applying them to these exact images. For any set of initial values of the Fourier coefficients chosen, the exact wave was reproduced.

Next, the method was applied to Poisson distributed image observations with the corresponding exact image values as expectations. Fig. 2 shows that the average over the period of these expectations is approximately equal to nine. This implies that the observations are very noisy since a Poisson variable with an expectation equal to nine has a standard deviation equal to three. This is clearly visible in the second row of Fig. 2 where the used realization of both images is shown. The real and imaginary parts of the wave reconstructed from these noisy images are shown in the second column of Fig. 1. The differences of the exact and these reconstructed real and imaginary parts are shown in the third column of Fig. 1.

The conclusion from this numerical experiment is that, in spite of the high noise level, the wave's most important

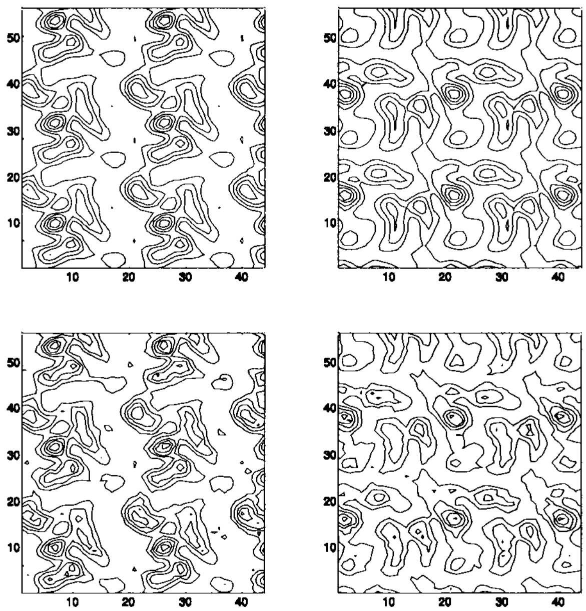

Fig. 3. Simulation experiment #2. Contours of exact (top) and Poisson distributed (bottom) intensities.

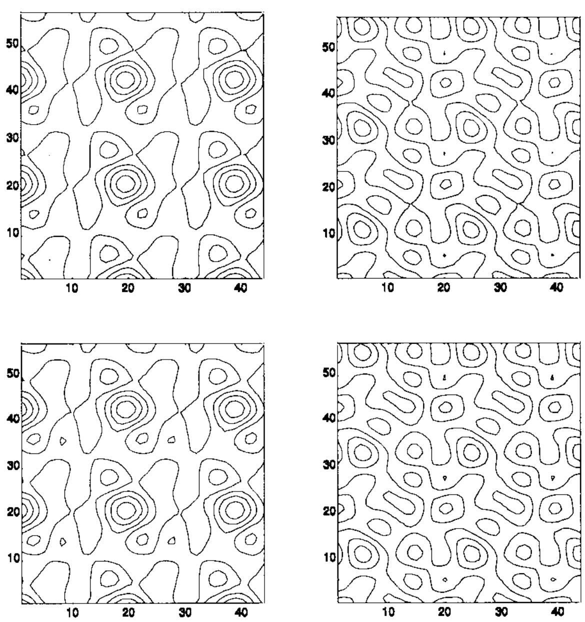

Fig. 4. Simulation experiment #2. Contours of real and imaginary parts of exact (top) and reconstructed (bottom) exit wave.

features, such as the location of its maxima and minima and their approximate relative heights, are very well reconstructed. Repeated experiments show that this is nearly always the case. In addition, the least-squares estimates of the Fourier coefficients have subsequently been used as initial values for maximizing the Poisson likelihood function (2). This results in slightly modified Fourier coefficient estimates having a smaller mean square error such as theory predicts.

B. Simulation Experiment #2

In this experiment, the wave and the observations are two-dimensional. The number of images is again equal to two. The number of pixels in either image is 44 × 57. The observations are Poisson distributed. The values of the transfer function are generated in the same way as in simulation experiment #1. The Fourier coefficients are shown in Table II. The first column and the first row describe k and ℓ, the harmonic numbers in the x and the y direction, respectively. The remaining entries are the complex Fourier coefficients for the (k, ℓ)th harmonic concerned. There are 24 complex Fourier coefficients and one real one, that is, 49 real parameters. This unrealistically large number of parameters has been chosen to put the method to the test. The Fourier coefficients actually used in the simulations are obtained by multiplying those of Table II by a scale factor of 0.6. This results in an average of the exact images of approximately 40, which for Poisson variates corresponds to a standard deviation of about 6.3. The numbers of periods observed are 2.3 and 2.6 in the x and y direction, respectively, and, since the number of pixels is 44 × 57, the number of pixels per period in these directions is 19.3 and 21.8, respectively.

TABLE II
RELATIVE AMPLITUDES OF FOURIER COEFFICIENTS IN EXPERIMENT #2

	- 2	- 1	0	1	2
- 2	1 + j	- 2- j	2 + j	3 - j	j
- 1	2 + 2j	3 + 2j	1.5 + j	3	1 + 0.5j
0	1 - j	1 + 3j	2	2 + j	2.5 - 1.5j
1	0.5 + 2j	0.5 + j	2 + j	j	1 + j
2	-1 + j	1	2 + 1.5j	1 - j	j

The minimizing of the least-squares criterion is carried out using the Levenberg–Marquardt method [9]. In spite of the relatively large number of parameters, no convergence problems occurred. The first row of Fig. 3 shows the contours of the exact images for both defoci. The second row shows a realization of these contours if the pixels are Poisson distributed with the corresponding pixels of the exact images as expectations. The influence of the noise is clearly visible. The first row of Fig. 4 shows the contours of the exact real and imaginary parts of the underlying wave, respectively. The second row of the same figure shows the contours of the corresponding real and imaginary parts of the wave reconstructed from the Poisson distributed intensities of Fig. 3. The differences of the contours of the exact real and imaginary parts and those of their reconstructed counterparts are hardly visible showing the quality of the reconstruction. Therefore, it would be unlikely that, in practice, the interpretation of the reconstructed wave would differ very much from that of the exact one.

VI. DISCUSSION AND CONCLUSIONS

A parameter estimation based method for reconstruction of spatially periodic specimen exit waves in transmission electron microscopes has been developed. Since the estimator employed is a maximum likelihood estimator, it is asymptotically most precise. As a result of the special attention paid to the convergence properties of the numerical nonlinear optimization required, the estimator has, in all simulations carried out, been found to converge to the optimal solution. Currently, an extension with optional estimation of the spatial periods is studied.

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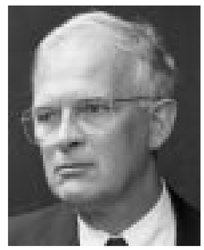

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