NIH Public Access
Author Manuscript
Ultramicroscopy. Author manuscript; available in PMC 2015 March 01.

Published in final edited form as:
Ultramicroscopy. 2014 March ; 138: 1–3. doi:10.1016/j.ultramic.2013.11.009.

GENERALIZATION OF THE MATSUMOTO-TONOMURA APPROXIMATION FOR THE PHASE SHIFT WITHIN AN OPEN APERTURE

Robert M. Glaeser and
Life Sciences Division, Lawrence Berkeley National Laboratory, University of California, Berkeley, CA 94720

Holger Müller
Physics Department, University of California, Berkeley, CA 94720

Abstract

As shown by Matsumoto and Tonomura, the phase shift imposed on an electron beam by an electrostatic phase plate is constant for all (straight) electron trajectories passing through a circular aperture, provided that (1) the electric field goes to zero at distances far above and below the aperture, and (2) the value of the phase shift at the boundary (i.e. perimeter of the aperture) is constant (Matsumoto and Tonomura, 1996). We now point out that the result can be valid for any shape of the hole in the aperture, and, furthermore, it requires only that the electric field is equal and opposite at large distances above and below the aperture, respectively. We also point out that the conditions of validity of the Matsumoto-Tonomura approximation constrain the phase shift across the open aperture to a quadratic algebraic form when the phase shift is not constant around the perimeter. Finally, it follows that the projection approximation for calculating the phase shift must fail for strong phase shifts of higher than quadratic form. These extensions of the original result of Matsumoto and Tonomura give further insight to the analysis of charging phenomena observed with apertures that are designed to produce contrast in in-focus images of weak phase objects.

Keywords

Aperture; charging; phase contrast

1. INTRODUCTION

A variety of aperture designs are currently under investigation that could, in theory, provide high contrast for weak phase objects under conditions of in-focus imaging (Glaeser, 2013). Some of these designs have shown excellent performance in proof-of-concept experiments. Nevertheless, the fabrication of well-performing devices has proven to be inconsistent, and even the best devices have had only a short working lifetime. The main difficulty

Corresponding author: Robert M. Glaeser, Life Sciences Division, Lawrence Berkeley National Laboratory, University of California, Berkeley, CA 94720, Phone: 510-642-2905, FAX: 510-486-6488, rmglaeser@lbl.gov.

NIH-PA Author Manuscript

NIH-PA Author Manuscript

encountered with such apertures is that they develop unwanted electrostatic charging, which in turn introduces unwanted phase shifts for electrons passing through the aperture. As a result, the contrast-transfer function has a much more complicated, even unknowable form than that expected from the design of the aperture.

We here revisit the mathematical constraints that limit the variation of the phase shift across the open area of an aperture, a question that was previously analyzed by Matsumoto and Tonomura (Matsumoto and Tonomura, 1996). While these authors were interested in the conditions required for the phase shift to be constant, we are now interested in the more general case when the phase shift is not constant across the aperture. We show that, if and only if the weak-lens approximation (see below) remains valid, charging can cause only an image shift along with additional defocus and astigmatism. As a result, it follows that electron trajectories through the aperture can no longer be described as straight lines if charging of the aperture introduces a more complicated phase distortion.

2. THEORY

Within what may be called the "weak-lens" approximation, the phase shift across an aperture is approximated by a function that satisfies a two-dimensional Laplace equation (Matsumoto and Tonomura, 1996):

$$\nabla^2_{x,y}\Psi(u,v)=0, \quad \text{Equation 1}$$

where $\Psi(u, v)$ is the phase shift for a ray passing through the point (u, v) within the open area of the aperture. The weak-lens approximation assumes that the phase shift is proportional to the integral of the electrostatic potential along a straight line (i.e. proportional to a 2-D projection of the 3-D electrostatic potential). In their derivation of Equation 1, Matsumoto and Tonomura also assumed that the electric field vanishes at large distances above and below the aperture. We note that their result also remains valid for the somewhat stronger condition that the value of the electric field at large distances is equal and opposite at distances far above and below the aperture.

One can see by substitution that $\Psi(u, v) = \Psi_0$, where Ψ_0 is a constant, is the (unique) solution of Equation 1 when $\Psi_{boundary}(u, v) = \Psi_0$. An aperture will generally be biased due to the contact potential, of course, since the aperture and its surroundings are usually made of dissimilar materials. As a result, this boundary condition is valid only when the electrostatic potential is axially symmetric about the center of the aperture.

When $\Psi_{boundary}(u, v)$ is not constant, however, one can again see by substitution that the solution of Equation 1 is of the form:

$$\Psi(u,v)=\Psi_0+a(u-u_0)+b(v-v_0)+c((u-u_0)^2-(v-v_0)^2)+d(u-u_0)(v-v_0), \quad \text{Equation 2}$$

where the constants *a, b, c,* and *d* can take any value. Note that this solution is valid regardless of the shape of boundary, as it does not assume that the variables are separable. The expression is written in a form that facilitates discussion of what to expect when the origin of the coordinate system used to represent the two-dimensional phase shift is offset by (u_0, v_0) relative to the origin of the electron diffraction pattern.

We also point out that the phase shift across a thin-film phase plate can be approximated by a solution of the two-dimensional Laplace equation, even though there is a hole in the center of the thin film, i.e. when there is a discontinuity in the phase shift at the edge of the hole. It is clear that the phase shift will still satisfy $\nabla^2_{x,y}\Psi(u,v)=0$ everywhere in the plane of the aperture except at the discontinuity. This is because the three-dimensional electrostatic potential still satisfies the homogeneous Laplace equation, as opposed to the inhomogeneous Poisson equation, as long as the thin-film phase plate itself is not charging. The analysis used by Matsumoto and Tonomura thus remains unchanged everywhere except at the boundary of the hole, where the phase shift is discontinuous and thus the Laplacian of the phase shift is undefined. We argue, however, that the phase shift across a physically realizable edge will be symmetrical and differentiable. We also argue that the Laplacian of the phase shift will be conservative, i.e. it will have equal and opposite values on the two sides of the edge. Thus, in the limit that the edge becomes more and more abrupt, the Laplacian of the phase shift will also be zero around the perimeter of the hole in the thin film.

3. DISCUSSION

In order to produce contrast in an in-focus image of a phase object, one can use a device (i.e. an aperture) that intentionally modifies either the amplitude or the phase (or both) of the scattered wave. Such a device can be represented mathematically by a generalized pupil function, as defined in chapter 6 of (Goodman, 1968), which is of the form:

$$\boldsymbol{P}(u,v)=P(u,v)e^{-i\Psi(u,v)} \quad \text{Equation 3}$$

where $P(u, v)$ is the (amplitude) transmittance of the aperture; $\Psi(u, v)$ is the phase shift (if any) applied by the aperture, whether intentionally or not; and bold-face font is used to designate a complex-valued function. In general, the aperture can have an arbitrary shape, as is shown schematically in Figure 1, and the unscattered beam can be located anywhere within the open area of the aperture. After the scattered wave function has been multiplied by such a generalized pupil function, the corresponding image wave function is the convolution product between the Fourier transform of the generalized pupil function and the image wave function that is produced without the aperture.

For simplicity, we assume that the specimen is a weak-phase object. We also ignore the effect of spherical aberration, and we assume that the microscope is properly stigmated and set to be in focus before an aperture is inserted. The image wave function produced without the aperture is thus modeled as $[1+ i\eta (x, y)]$, where $\eta (x, y)$ represents the spatial modulation of the phase of the electron wave that is transmitted through the specimen. The scattered wave function, in turn, is modeled as $\delta (u, v) + i\boldsymbol{F}(u, v)$, where $\boldsymbol{F}(u, v)$ is the Fourier transform of $\eta (x, y)$.

3.1. Physical consequences of each term in Equation 2

In this section of the discussion we assume that a phase shift, if any, is “locally” constant across the unscattered beam. Our purpose in adopting this simplification is to discuss the consequences when a variable phase shift, of the form in Equation 2, is applied the scattered wave function.

Ψ_0, the constant phase shift in Equation 2, is also known as a “piston” aberration. A phase shift that is constant across the entire aperture is unimportant because it produces no observable effect in the image intensity.

The bi-linear term in Equation 2 applies a linear phase ramp across the scattered wave function. When an inverse Fourier transform then converts this modified scattered-wave function to an image-wave function, the linear phase ramp causes only an image shift. Image shift will seldom be objectionable, even if the amount and direction of shift changes as the aperture is moved relative to the electron diffraction pattern.

The quadratic terms in Equation 2 introduce defocus and astigmatism, however, which again change as the aperture is moved relative to the electron diffraction pattern. It may be argued that the image defocus and astigmatism already have to be compensated by the microscope operator each time that the specimen is moved to a new area, because of uncontrolled changes in specimen height and (possible) charging of the specimen. Nevertheless, these quadratic terms will cause unwanted complications if the phase-contrast aperture has to be moved frequently during data collection. It would thus be better it they were avoided altogether by ensuring that the phase shift is constant around the boundary of the aperture.

3.2. Focus and astigmatism changes are an indication of charging of the phase-contrast aperture

Experimentally, we often observe changes in astigmatism and focus as the aperture is moved relative to the diffraction pattern. From Equation 2 and the subsequent discussion, it must also be that the phase shift at the perimeter is not constant. In other words, we can conclude that there is a variable amount of charging around the perimeter. One can distinguish three scenarios that account for the amount of charging not being constant around the perimeter of an aperture.

In the first scenario, it is imagined that the aperture is biased relative to the surrounding material, as mentioned above. If the electrostatic potential between the aperture and the surrounding material is not symmetrical about the center of the aperture, then the projection of the potential onto the plane of the aperture will not be constant around the perimeter.

In the second scenario, a fixed pattern of variable phase may exist around the perimeter of the aperture, and this pattern might be different from one aperture to the next. This could be due, for example, to work-function differences or other “patch potential” effects around the perimeter. In this scenario, as in the first one, the aperture would exhibit “charging effects”, but these effects would be present even in the absence of scattered electrons hitting the aperture.

In the third scenario, we group all cases where the amount of charging does depend upon the intensity of electron irradiation at different parts of the aperture. One example might be the build-up of a positive charge on any insulating material on the surface of the aperture. Another example might be changes in work function or in contact potential due to radiation-induced structural changes in the most intensely irradiated area of the aperture.

3.3. Beam deflection must be invoked in order to understand anomalous behavior of the CTF that is due to charging of the aperture

In many cases the phase shifts observed experimentally in the contrast transfer function (the CTF) cannot be accounted for by the quadratic polynomial written in Equation 2. An extreme example is shown in Figure 5 of (Glaeser et al., 2013), where the FFT of an image shows concentric oscillations of contrast that are centered at the point where the focused electron beam had been intentionally allowed to touch the edge of the aperture. Another extreme example is shown in Figure 5 of (Marko et al., 2011), where the FFT of an image shows a radial dependence that is strongly different from what can be accounted for by defocus, astigmatism, and spherical aberration. In cases like these, one must conclude that the phase shifts, at least in some areas of the aperture, are not correctly accounted for as line integrals of the electrostatic potential; in other words, the electron trajectories must not be straight lines and the weak lens approximation is not valid.

Acknowledgments

This work was supported in part by NIH grant GM083039 and by NSF award #029907-002.

References

Glaeser RM. Invited Review Article: Methods for imaging weak-phase objects in electron microscopy. Review of Scientific Instruments. 2013; 84 In Press.

Glaeser RM, Sassolini S, Cambie R, Jin J, Cabrini S, Schmid AK, Danev R, Buijsse B, Csencsits R, Downing KH, Larson DM, Typke D, Han BG. Minimizing electrostatic charging of an aperture used to produce in-focus phase contrast in the TEM. Ultramicroscopy. 2013; 135:6–15. [PubMed: 23872037]

Goodman, JW. Introduction to Fourier optics. San Francisco: McGraw-Hill; 1968.

Marko M, Leith A, Hsieh C, Danev R. Retrofit implementation of Zernike phase plate imaging for cryo-TEM. Journal of Structural Biology. 2011; 174:400–412. [PubMed: 21272647]

Matsumoto T, Tonomura A. The phase constancy of electron waves traveling through Boersch's electrostatic phase plate. Ultramicroscopy. 1996; 63:5–10.

- The phase of an electron wave front is affected by charging of the aperture.
- The phase is constrained to a quadratic form if trajectories are straight lines.
- The projection approximation must fail if the distortion is of higher order.

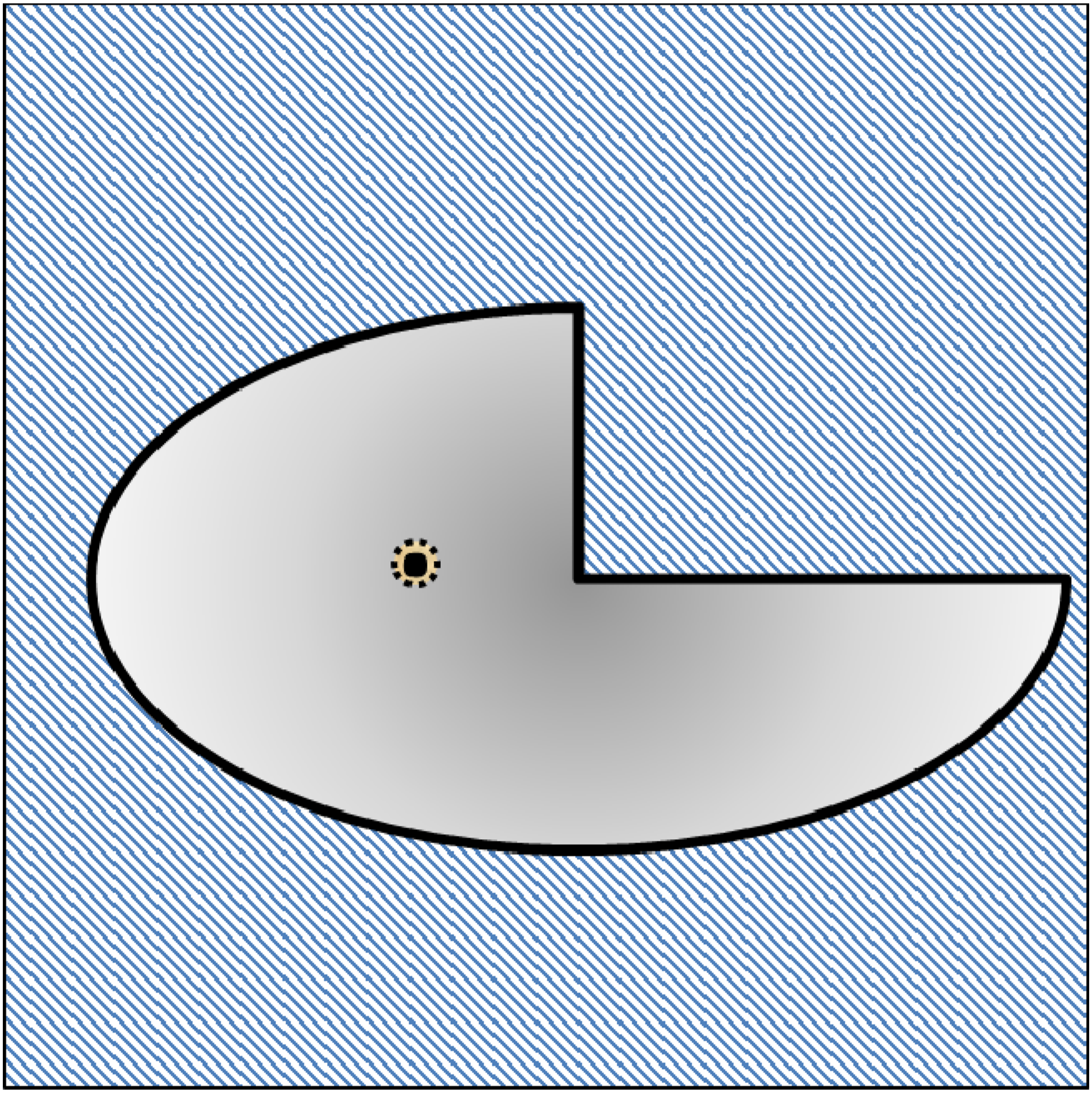

Figure 1.

Schematic representation of a generalized (i.e. complex-valued) pupil function like that referred to in Equation 3. The clear area in this figure, which need not be symmetrical in shape, represents the open area of the aperture, and the hatched area in the figure represents the opaque area. The continuous shading within the clear area represents the possibility that the phase of the generalized pupil function may vary across the open area. The small, dashed circle with a dot in the center represents the location of the focused, unscattered beam of the electron diffraction pattern, which can be placed at an arbitrary location within the open area of the aperture.