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Quantitative electric field mapping in thin specimens using a segmented detector: Revisiting the transfer function for differential phase contrast

Takehito Seki [a], Gabriel Sánchez-Santolino [a], Ryo Ishikawa [a], Scott D. Findlay [b], Yuichi Ikuhara [a,c], Naoya Shibata [a,c,*]

[a] Institute of Engineering Innovation, School of Engineering, The University of Tokyo, Yayoi 2-11-16, Bunkyo-ku, Tokyo 113-8656, Japan
[b] School of Physics and Astronomy, Monash University, Clayton, Victoria 3800, Australia
[c] Nanostructure Research Laboratory, Japan Fine Ceramic Center, 2-4-1 Mutsuno, Atsuta-ku, Nagoya 456-8587, Japan

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ABSTRACT

Differential phase contrast in scanning transmission electron microscopy can visualize local electromagnetic fields inside specimens. The contrast derived from first moments, the so-called center of mass, of the diffraction patterns for each probe position can be quantitatively related to the local electromagnetic field under the phase object approximation. While only approximate first moments can be obtained with a segmented detector, in weak phase objects the fields can be accurately quantified on the basis of a phase contrast transfer function. Through systematic image simulations we further show that the quantification based on the approximated first moment is a good approximation also for strong phase objects.

1. Introduction

Nano-scale spatial distribution of electromagnetic fields inside materials and devices is of critical importance to fundamentally understand the origins of their functional properties. Scanning transmission electron microscopy (STEM) boosted by aberration-correction technology enables us not only to directly observe atomic-scale local structures such as interfaces and surfaces of materials, but also to directly observe the local electromagnetic field structures they induce. In ordinary STEM, the focused electron probe is scanned across the specimen, and STEM images are formed by using annular type dark-field detectors to collect electrons scattered through high angles as a function of probe position. On the other hand, electrons transmitted through the specimen will be deflected by the electromagnetic fields inside it. The deflection is measurable by subtracting signals detected in diametrically opposing segments in an azimuthally segmented detector. The idea of enhancing phase contrast by subtracting signals from different detectors in STEM was first proposed by Rose [1]. Dekker and de Lang showed that the contrast obtained by subtracting signals from split detectors is related to the gradient of the phase of the specimen transmission function (and thus to the electromagnetic fields), and this imaging mode is called differential phase contrast (DPC) [2]. Thus, DPC STEM can, in principle, visualize electromagnetic fields inside materials in real space. Phase contrast transfer functions (PCTFs) of DPC imaging in the one-dimensional and two-dimensional cases were given by Dekker and de Lang [2] and by Rose [3], respectively. These mathematical descriptions are valid within the weak phase object approximation (WPOA). The linear approximation was extended to strong phase objects by Waddell and Chapman [4]. They showed that the first moment, or center of mass, of the electron intensity distribution in the detector plane for each probe position is closely related to the difference signals, and that a 'first-moment detector' gives a PCTF valid under the phase object approximation (POA). Since that early work, PCTFs for segmented and first-moment detectors have been discussed in detail [5–12]. Recently, the first-moment measurement was interpreted more simply according to the Ehrenfest's theorem [13]. While a segmented detector gives only an approximation to the actual first moment, approximating the first moment using a multi-segmented detector has been shown to retrieve the strong phase more accurately than the conventional WPOA method based on the PCTF for all but the thinnest (a few nanometers or less) of crystals [14]. Very recently, we proposed a simple quantification method based on the PCTF for the approximated first moment [15], showing the combination of the approximated first moment and the PCTF methods can be more accurate than using each method independently.

* Corresponding author at: Institute of Engineering Innovation, School of Engineering, The University of Tokyo, Yayoi 2-11-16, Bunkyo-ku, Tokyo 113-8656, Japan.
E-mail address: shibata@sigma.t.u-tokyo.ac.jp (N. Shibata).

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Experimentally, DPC imaging has been performed mainly using segmented detectors for visualizing electromagnetic fields inside materials: e.g. magnetic domains [16–18], ferroelectric domains [19], p-n junctions [20], quantum wells [21], skyrmions [22,23] and atomic electric fields in crystals [19]. Recently, the first-moment detector was realized using a pixelated detector for atomic resolution DPC imaging [13]. Though segmented detectors yield only an approximation to the first moment, scintillator-type segmented detectors are still two or three orders of magnitude faster than the state-of-the-art pixelated detectors. The fast scan rate is of great advantage for atomic resolution imaging because (1) images from larger areas can be obtained, (2) electron irradiation damage and contamination of the sample can be suppressed, and (3) the probe positions are more accurate for post-processing (e.g. for integration and divergence of the measured fields to obtain potential and charge-density information, respectively [11]).

In this paper, we aim to further develop a better quantification method for DPC signals obtained using a multi-segmented detector by combining the approximated first moment and the PCTF methods. The PCTF formula given by Rose [3] is re-derived to make clear how to quantify the electromagnetic fields via PCTFs. We calculate several PCTFs for a 16-segment detector [24] on the basis of the approximated-first-moment method. We show this method is more accurate for strong phase objects than the conventional PCTF for simply subtracted segmented detector signals. Finally, we explore the validity of the PCTF description based on the approximated first moment through detailed image simulation.

2. Theory

In the early work by Rose [3], the PCTF for coherent STEM imaging was derived in the coordinates of the equivalent TEM optical system based on the reciprocity theorem [25]. Here, the PCTF is re-derived directly in the coordinates of the STEM optical system.

We adopt a coordinate system in which $\boldsymbol{r}_\perp$ denotes the real space coordinate in the plane perpendicular to the optical axis and $\boldsymbol{k}_\perp$ denotes the reciprocal space coordinate conjugate to $\boldsymbol{r}_\perp$. The wave function of a STEM probe located at $\boldsymbol{r}_\perp = \boldsymbol{R}$ can be written as [26]

$$\psi_{\text{in}}(\boldsymbol{r}_\perp, \boldsymbol{R}) = \int T(\boldsymbol{k}_\perp) e^{2\pi \text{i}\boldsymbol{k}_\perp \cdot (\boldsymbol{r}_\perp - \boldsymbol{R})} \text{d}\boldsymbol{k}_\perp \quad (1)$$

$$T(\boldsymbol{k}_\perp) = A(\boldsymbol{k}_\perp)\exp(-\text{i}\chi(\boldsymbol{k}_\perp)) \quad (2)$$

where $T(\boldsymbol{k}_\perp)$ is the lens transfer function and $\chi(\boldsymbol{k}_\perp)$ is the lens aberration function. The aperture function $A(\boldsymbol{k}_\perp)$ is uniform within the aperture, zero beyond it, and normalized such that the total intensity is unity. Under the WPOA, the wave function at the diffraction plane after transmitting a thin specimen of projected potential $v_{\text{prj}}(\boldsymbol{r})$ is written as

$$\tilde{\psi}_{\text{out}}(\boldsymbol{k}_\perp, \boldsymbol{R}) = \int \left(1 + \text{i}\sigma v_{\text{prj}}(\boldsymbol{r}_\perp)\right) \psi_{\text{in}}(\boldsymbol{r}_\perp, \boldsymbol{R}) e^{-2\pi \text{i}\boldsymbol{k}_\perp \cdot \boldsymbol{r}_\perp} \, \text{d}\boldsymbol{r}_\perp \quad (3)$$

where $\sigma = 2\pi me\lambda/h^2$ is an interaction constant. m, e (>0), λ and h denote the electron's relativistic mass, the elementary charge, the electron's wave length and Planck's constant, respectively. For simplicity we only consider electric fields, but the method can readily be generalized to include the vector potential if the sample is magnetic [27]. The measured diffraction pattern is the intensity of this wave function. The Fourier transform of the diffraction pattern intensity with respect to coordinate $\boldsymbol{R}$ can be shown to be

$$\begin{aligned} \tilde{I}(\boldsymbol{k}_\perp, \boldsymbol{K}) &= A(\boldsymbol{k}_\perp)\delta(\boldsymbol{K}) \\ &+ \text{i}\sigma V_{\text{prj}}(\boldsymbol{K})[T^*(\boldsymbol{k}_\perp)T(\boldsymbol{k}_\perp - \boldsymbol{K}) - T(\boldsymbol{k}_\perp)T^*(\boldsymbol{k}_\perp + \boldsymbol{K})] \end{aligned} \quad (4)$$

where $V_{\text{prj}}(\boldsymbol{K})$ is the Fourier transform of $v_{\text{prj}}(\boldsymbol{R})$, and absorption (i.e. any imaginary component of the real space potential) is ignored. The Fourier transform of the intensity in a general coherent STEM image is given by $\mathcal{F}[I_{\text{STEM}}(\boldsymbol{R})] = \int \tilde{I}(\boldsymbol{k}_\perp, \boldsymbol{K}) D(\boldsymbol{k}_\perp) \text{d}\boldsymbol{k}_\perp$, where $\mathcal{F}$ denotes Fourier transform with respect to $\boldsymbol{R}$ and $D(\boldsymbol{k}_\perp)$ denotes a detector response function. Thus, the PCTF $\beta(\boldsymbol{K})$ may be defined as

$$\mathcal{F}[I_{\text{STEM}}(\boldsymbol{R})] = \delta(\boldsymbol{K}) \int A(\boldsymbol{k}_\perp) D(\boldsymbol{k}_\perp) \text{d}\boldsymbol{k}_\perp + \sigma V_{\text{prj}}(\boldsymbol{K}) \beta(\boldsymbol{K}) \quad (5)$$

$$\begin{aligned} \beta(\boldsymbol{K}) = \text{i} \int & A(\boldsymbol{k}_\perp) D(\boldsymbol{k}_\perp) [A(\boldsymbol{k}_\perp - \boldsymbol{K}) \exp(-\text{i}\chi(\boldsymbol{k}_\perp - \boldsymbol{K}) + \text{i}\chi(\boldsymbol{k}_\perp)) \\ & - A(\boldsymbol{k}_\perp + \boldsymbol{K}) \exp(\text{i}\chi(\boldsymbol{k}_\perp + \boldsymbol{K}) - \text{i}\chi(\boldsymbol{k}_\perp))] \text{d}\boldsymbol{k}_\perp \end{aligned} \quad (6)$$

The integral in Eq. (5) is zero if the detector response function is antisymmetric ($D(-\boldsymbol{k}_\perp) = -D(\boldsymbol{k}_\perp)$). Detector response functions for DPC imaging normally satisfy this condition, as will be discussed below.

According to Ehrenfest's theorem [28], the expectation value of the momentum transferred to the electron in transmitting a thin specimen, namely the first moment of the intensity of the diffraction pattern, is proportional to the weighted average of the projected electric field inside the probe as described below [11,13,27,29]

$$\int \boldsymbol{E}_{\perp\text{prj}}(\boldsymbol{r}_\perp) \left|\psi_{\text{in}}(\boldsymbol{r}_\perp, \boldsymbol{R})\right|^2 \text{d}\boldsymbol{r}_\perp = -\frac{h\nu}{e} \int \boldsymbol{k}_\perp \left|\tilde{\psi}_{\text{out}}(\boldsymbol{k}_\perp, \boldsymbol{R})\right|^2 \text{d}\boldsymbol{k}_\perp \quad (7)$$

where the projected field $\boldsymbol{E}_{\perp\text{prj}}(\boldsymbol{r}_\perp)$ is defined as the integration along the optical axis of electric field component perpendicular to the optical axis: $\boldsymbol{E}_{\perp\text{prj}}(\boldsymbol{r}_\perp) = \int \boldsymbol{E}_\perp(\boldsymbol{r}_\perp, z)\text{d}z$. This measurement is possible using a first-moment detector, which can be experimentally realized using a pixelated detector with a detector response function $D_\alpha^{FM}(\boldsymbol{k}_\perp) = k_\alpha$, where α denotes x or y, and k_α denotes the α component of $\boldsymbol{k}_\perp$. Applying this method, the measured field $\bar{E}_{\alpha,\text{prj}}^{\text{FM}}(\boldsymbol{r}_\perp)$ is given by

$$\bar{E}_{\alpha,\text{prj}}^{\text{FM}}(\boldsymbol{R}) \overset{\text{def}}{=} -\frac{h\nu}{e} I_{\text{DPC},\alpha}^{\text{FM}}(\boldsymbol{R}) = E_{\alpha,\text{prj}}(\boldsymbol{R}) \otimes \left|\psi_{\text{in}}(-\boldsymbol{R}, \boldsymbol{0})\right|^2 \quad (8)$$

where $I_{\text{DPC},\alpha}^{\text{FM}}(\boldsymbol{R}) = \int \tilde{I}(\boldsymbol{k}_\perp, \boldsymbol{R}) D_\alpha^{\text{FM}}(\boldsymbol{k}_\perp) d\boldsymbol{k}_\perp$ is the DPC image obtained by the first-moment detector and $\otimes$ denotes convolution. Here, the measured field is equal to the true field convolved with the probe intensity function. This equation holds not only within the WPOA but also within the phase object approximation (POA) [4,13]. Using a segmented detector, the first moment of the diffraction pattern can be approximated using the following detector response function [14]

$$D_\alpha^{\text{aFM}}(\boldsymbol{k}_\perp) = \begin{cases} k_{\alpha,j}^{\text{CoM}} & \text{if } \boldsymbol{k}_\perp \text{ lies within the } j\text{th segment,} \\ 0 & \text{otherwise,} \end{cases} \quad (9)$$

where $k_{\alpha,j}^{\text{CoM}}$ denotes the α component of the geometrical center-of-mass of the jth detector segment. The DPC image signal using a segmented detector can thus be evaluated as $I_{\text{DPC},\alpha}^{\text{aFM}}(\boldsymbol{R}) = \sum_j k_{\alpha,j}^{\text{CoM}} \tilde{I}_j$, where $\tilde{I}_j$ denotes the intensity detected in the jth segment (which, for consistency with the normalization of the aperture function, should be normalized by the total intensity of the bright field disk in the absence of the specimen). If the segment arrangement is symmetrical with respect to the origin of $\boldsymbol{k}_\perp$, the detector function is antisymmetric and the first term in Eq. (5) should be zero. The 16-segment detector and the centers-of-mass of each segment are shown in Fig. 1(a).

Under the WPOA, the DPC image signal can be described as

$$I_{\text{DPC},\alpha}(\boldsymbol{R}) = \mathcal{F}^{-1}\left[\sigma V_{\text{prj}}(\boldsymbol{K}) \beta_\alpha(\boldsymbol{K})\right], \quad (10)$$

where $\beta_\alpha(\boldsymbol{K})$ is the PCTF obtained in Eq. (6) using $D_\alpha^{\text{FM}}(\boldsymbol{k}_\perp)$ or $D_\alpha^{\text{aFM}}(\boldsymbol{k}_\perp)$, depending on whether a pixelated or segmented detector is used. $\beta_\alpha(\boldsymbol{K})$ is purely imaginary when $D(-\boldsymbol{k}_\perp) = -D(\boldsymbol{k}_\perp)$