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Prospects of annular differential phase contrast applied for optical sectioning in STEM☆

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ABSTRACT

The annular differential phase contrast (ADPC) mode in a third-order spherical aberration-corrected scanning transmission electron microscope (STEM) has recently been realized at an operating voltage of 300 kV by inserting a physical Fresnel phase plate in front of the objective lens and by using a detector geometry which matches that of the Fresnel phase plate [1]. By image calculation we explore the feasibility of this mode for the voltage range of 20–80kV. Alternatively, we mimic the Fresnel phase plate material-free with the help of the adjustable aberrations of the corrector. The additional correction of chromatic aberration, fifth-order spherical aberration and image spread improves significantly the resolution and contrast. Under these advanced conditions it is possible to achieve optical sectioning in the ADPC mode with atomic resolution and a depth of field shorter than 3 Å for an accelerating voltage of 30 kV. Moreover, we show that the contrast obtained in the ADPC mode is clearly superior over the contrast in incoherent bright-field (IBF) and high-angle annular dark-field (HAADF), the two other common methods in STEM. We propose that with the advanced ADPC method applied in STEM, the investigation of the inner structure of thick samples will be possible without slicing.

1. Introduction

Scanning transmission electron microscopy (STEM) usually employs the high-angle annular dark-field (HAADF) mode for visualizing the atomic structure of objects. However, HAADF images show primarily heavy atoms because the contrast is approximately proportional to Z^2. Aberration correction offers the possibility to visualize low-Z atoms in the STEM by employing differential phase contrast (DPC) methods [2–5]. Two representative procedures are the annular differential phase contrast (ADPC) mode and the integrated differential phase contrast (IDPC) mode.

The ADPC mode uses a Fresnel phase plate, which is located at the front-focal plane of the objective lens. The phase plate can be realized either by adjusting the defocus and the correctable spherical aberrations appropriately [2,4] or by introducing a material phase plate [1]. The introduction of the Fresnel phase plate results in constructive and destructive interference patterns on the detector, and the interference patterns depend strongly on the geometry of the phase plate. The intensity resulting from the constructive and destructive interference is collected separately by the segments of a ring detector. The difference of these signals removes the nonlinear information and enhances the phase contrast [4]. This method was given the name MIDI–STEM [1], where MIDI stands for 'matched illumination and detector interferometry'. Considering that this method belongs to the DPC techniques, we decide to replace the name MIDI with ADPC, where A stands for annular, indicating that the geometry of the illumination and detector is different from the ones applied by other DPC techniques.

In the IDPC mode, the DPC images are first acquired by subtracting the signals collected by opposite segments of a quadrant detector [4]. By integrating the difference signals along the two symmetric axes of the segments, IDPC is obtained, demonstrating its advantage by suppressing high-frequency noise [4,6].

Besides these two procedures employing either a ring detector or a quadrant detector, other methods are proposed, which utilize different detector geometries and different algorithms for combining the signals of the detector elements. The proposed detector geometries comprise the bisectioned detector [3], the annular quadrant detector [7,8], the 16-segments detector [9] and the unitary detector [10]. Moreover, the pixelated detector is also used for DPC imaging since it can be used to generate any detector geometry with high efficiency and flexibility [11–13].

DPC methods in STEM have been used not only for visualizing low-Z materials [1,14], but have also been applied to investigate the magnetic structure [7,13,15], or the electronic structure [16–19] of the sample. An in-depth review of the DPC methods can be found in [20].

Among the different STEM–DPC techniques, we restrict our

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investigation to the ADPC method, which employs a Fresnel phase plate and a bright-field ring detector [1,2,4]. The development of the C_c/C_n-corrector (n=3, 5) offers the possibility to fully utilize all differential phase contrast procedures [5]. Chromatic aberration is caused by the energy spread of the incident electrons, which reduces resolution and contrast of the STEM images especially at low accelerating voltages. Our theoretical investigations on the ADPC method are dedicated to a low-voltage STEM, which employs the C_c/C_n-corrector (n = 3, 5) as a probe corrector [5,21]. We suggest a voltage range of 20–80 kV such as in the SALVE (sub-Angstroem low-voltage electron microscopy) instrument. The C_c/C_n-corrector (n = 3, 5) provides the possibility to generate a material-free Fresnel phase plate by adjusting the spherical aberration coefficients and the defocus appropriately [4]. Combined with a pixelated detector matching the geometry of the Fresnel phase plate, the C_c/C_n-corrected STEM–DPC (n = 3, 5) can achieve phase contrast with high flexibility and efficiency.

2. Method

The intensity pattern formed in the bright-field region of the STEM detector results from the interference of the elastically scattered wave with the non-scattered part of the incident wave. Optimum linear phase contrast is obtained in the aberration-corrected STEM by nullifying the chromatic aberration and by introducing a Fresnel phase plate. The Fresnel phase plate can be realized either by adjusting the spherical aberration coefficients C_n, n = 3, 5, ... and the defocus properly [4], or by employing a physical phase plate together with C_n = 0 [1]. The phase plate must be designed in such a way that the areas of constructive and destructive interference are equal. Fig. 1 illustrates the method on the left and the plots on the right show the real part and the imaginary part of the transfer function $T(\vec{q})$, as well as the interference pattern in the detector plane. The interference pattern has been calculated by assuming that the probe is centred on a single atom. The calculations confirm that the structure of the interference pattern is strongly related to the structure of the phase of the incident wave. Maximum phase contrast is obtained by recording the areas of constructive and destructive interference with separate detectors, and by subtracting subsequently one signal from the other. A mathematical derivation of the contrast obtained in the ADPC mode is given in Appendix A.

The contrast of weak phase objects is linearly related to the object potential in the ADPC mode. This behavior does not hold true for strong phase objects as outlined in Appendix A. For a weak phase object, the ADPC contrast can be expressed with a sufficient degree of accuracy by summing up the contributions of the individual atoms (A.12).

3. Influence of chromatic aberration and Johnson noise on the ADPC mode in STEM

In this section we will show the necessity to eliminate the chromatic aberration and to reduce the Johnson noise in order to successfully apply the suggested method. The effect of chromatic aberration on the contrast transfer function is taken into account by the mixed temporal coherence function:

$$E_c(\vec{q}, \vec{q}') = \exp[-\frac{1}{2}(\pi\sigma_c\lambda)^2(q^2 - q'^2)^2], \quad \sigma_c = C_c\frac{\sqrt{\langle(\Delta E)^2\rangle}}{E_0}. \tag{1}$$

The standard deviation of the chromatic focal spread σ_c depends on the coefficient C_c of the chromatic aberration, the mean quadratic energy spread $\langle(\Delta E)^2\rangle$, and the nominal electron energy E_0. The variables $\vec{q}$ and $\vec{q}'$ are 2D spatial frequency vectors, and $\lambda = 2\pi/k$ is the wavelength of the electron.

As discovered recently, the image spread results from the Johnson noise, which is caused by the thermally induced currents in the lenses and in the elements of the corrector and the vacuum tube [22]. Image spread limits appreciably the attainable resolution and contrast in the C_c/C_n-corrected microscope (n = 3, 5). The influence of the Johnson noise on the contrast transfer is described by the mixed image spread function $E_{is}(\vec{q}, \vec{q}')$ [23]:

$$E_{is}(\vec{q}, \vec{q}') = \exp[-2(\pi\sigma_\epsilon)^2(\vec{q} - \vec{q}')^2]. \tag{2}$$

The standard deviation σ_ϵ of the image spread depends on the voltage and is in the range between 20 pm and 30 pm for the SALVE microscope.

The contrast transfer function for the ADPC mode is given by

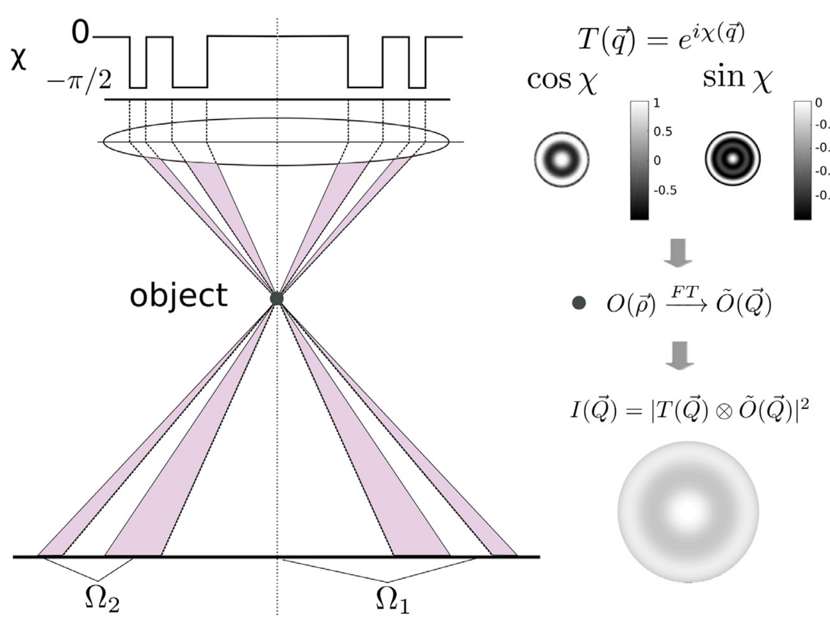

Fig. 1. Optimum differential phase contrast in the STEM requires a Fresnel phase plate, which must be designed in such a way that the area introducing a phase shift $-\pi/2$ equals that introducing no phase shift. This ideal phase plate can be approximately realized by adjusting the coefficients C_n (n = 1, 3, 5) appropriately. The simulations on the right illustrate the resulting phase transfer function $T(\vec{q})$ structured by a C_c/C_3–aberration corrector. The interference pattern $I(\vec{Q})$ shown on the bottom is obtained when the probe is centered on an atom.