Ultramicroscopy 114 (2012) 72–81

Contents lists available at SciVerse ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

Using a monochromator to improve the resolution in TEM to below 0.5 Å. Part I: Creating highly coherent monochromated illumination

P.C. Tiemeijer [a,*], M. Bischoff [a], B. Freitag [a], C. Kisielowski [b]

[a] FEI Company, P.O. Box 80066, 5600 KA Eindhoven, The Netherlands
[b] National Center for Electron Microscopy, Lawrence Berkeley National Laboratory, One Cyclotron Road, Berkeley, CA 94720, USA

ARTICLE INFO

Article history:
Received 7 August 2008
Received in revised form
15 December 2011
Accepted 15 January 2012
Available online 25 January 2012

Keywords:
TEM
Monochromator
Brightness

ABSTRACT

Chromatic aberration limits the resolution in spherical-aberration corrected Transmission Electron Microscopy to approximately 0.7 Å at 300 kV. The energy spread in the beam is the main contribution to the chromatic aberration. This spread can be reduced with a monochromator. Another limitation to the resolution in TEM can be the finite brightness of the source and the consequent partial spatial coherence of the illumination. This limitation becomes important when spherical aberration and/or defocus are present such as in uncorrected TEM or in focal-series reconstruction in TEM. We used a monochromator optimized for minimum brightness loss and a prototype 'high-brightness' gun, and obtained brightness *after* monochromation comparable to that of the standard Schottky FEG *before* monochromation. The images were acquired on the prototype TEAM 0.5 microscope, which was developed on a Titan platform by increasing its electrical and mechanical stability.

1. Introduction

The optical resolution in conventional Transmission Electron Microscopy (TEM) is limited by the two fundamental aberrations of the objective lens, spherical aberration and chromatic aberration. Spherical aberration blurs the image information beyond the Scherzer point resolution of the TEM. This blurred information can be retrieved with post-processing techniques such as holography [1] or focal-series reconstruction [2]. Since the development of Cs correctors [3], direct compensation of the spherical aberration (Cs) and higher order aberrations is increasingly applied. Today, post-processing and/or Cs correction can give directly interpretable TEM images down to 0.8…1.0 Å [4–8] in the mid-voltage range.

Chromatic aberration (Cc) determines the information limit of the TEM provided mechanical and electrical instabilities can be neglected. The energy spread in the beam causes the objective lens not to focus at a specific focal distance, but rather over an interval of focal distances. This interval is proportional to the chromatic aberration, and results in blurring of the image. The limit beyond which details are irretrievably lost by chromatic blurring is called the information limit. Cc correctors have been proposed [9–11] and built [12]. Until recently, the extreme demands on the electrical stability and mechanical accuracy posed by these correctors were beyond technical feasibility, and this has prevented improvement of the information limit by this method for a long time. An alternative way to improve the information limit is to monochromize the beam. It has been demonstrated that monochromation can improve the information limit from 1.1 Å to well below 1.0 Å on a 200 kV TEM [13], and recently, that monochromation can yield atomic resolution at high tensions as low as 20…50 kV [14-16].

However, the monochromator does reduce the brightness of the beam because it removes part of the energy spectrum, and it can further reduce brightness if aberrations and especially Coulomb interactions in the monochromator are blurring the image of the source. The first effect is inevitable, but the other effects have been suitably minimized in our design [17].

In the existing literature, little attention has been paid to quantifying the effect of beam brightness on resolution. It is well known that beam brightness determines how much current can be focused into a small probe for a given convergence angle. It is less well recognized that beam brightness also limits the current obtainable in a parallel TEM beam for a given coherence angle and, thereby the possible electron count per pixel. This correspondence between brightness in STEM and in TEM can easily be understood from the correspondence between the focused probe in STEM and the final cross-over in the illumination in TEM. The demands on brightness become especially strict when images are taken out-of-focus. Such defocus can be negligible when single images are taken in a Cs-corrected microscope, since a single image can be taken at a focus which is only a few nanometers

* Corresponding author. Tel.: +31 6 1208 9765; fax: +31 40 2356655.
E-mail address: p.tiemeijer@fei.com (P.C. Tiemeijer).

0304-3991/$ - see front matter © 2012 Elsevier B.V. All rights reserved.
doi:10.1016/j.ultramic.2012.01.008

away from Gaussian focus. However, larger defocus does occur in imaging of Life Science specimens or in focal series reconstruction. We will discuss the brightness requirements in out-of-focus imaging in detail in a companion paper which reports on our experiments on monochromated focal series reconstruction [18]. In that paper we derive that, for a given defocus and for a given brightness, the pixel count rate drops with the fourth power of the targeted resolution. Of course, a low count rate can sometimes be compensated by a longer exposure time, but this option is limited by radiation damage, specimen drift and other instabilities. Therefore it is essential to have maximal brightness if one aims at ultimate resolution in focal series reconstruction. In recent years, we have tested monochromators for improving the information limit in various microscope configurations. Indeed, we experienced that brightness is an essential parameter since only careful tuning of the condenser lenses and the monochromator results in a suitable balance between beam current and coherence angle that yields reasonable count rates and resolution improvement.

Monochromation can lower the information limit from about 0.7 Å to about 0.5 Å. In the absence of electronical and mechanical instabilities, an energy spread of 0.27 eV results in an information limit of 0.5 Å at 300 kV. However, for a long time, microscopes were not stable enough for demonstrating 0.5 Å. In the recent past, a prototype development for the TEAM (Transmission Electron Aberration-corrected Microscope) Project [19] was completed, and the instrument was optimized for 0.5 Å resolution. This microscope was based on an FEI Titan 80-300 instrument and equipped with CEOS designed Cs correctors on the probe forming side and on the imaging side. The whole column was installed in a thermal and acoustic enclosure to reduce sensitivity to the environment. It also included modifications to power supplies for noise reduction of the deflectors and the objective lens. The standard Schottky FEG was replaced by a prototype 'high-brightness' gun with a brightness comparable to that of the cold field emitter, but with the better emission stability and the lower emission noise of the Schottky FEG. The emission stability of this gun is beneficial for recording focal series. These modifications enabled, for the first time, information transfer to below 0.5 Å in TEM and STEM mode at 300 kV [20] and 1 Å in TEM mode at 80 kV [21-23]. We also succeeded for the first time in demonstrating the full benefit of the present monochromator gun assembly for high resolution TEM by showing that information to below 0.5 Å is transferred not only in single imaging but also in focal-series reconstructions [24]. These experiments were done during the final acceptance tests of the TEAM 0.5 microscope at the factory in August 2007. By now the microscope has been installed at the National Center for Electron Microscopy in Berkeley and is performing according to specifications.

In this paper we describe the optics that we use for creating high-brightness monochromated TEM illumination. We have chosen for a monochromator design with relatively simple optics and of relatively short optical length. The advantage of such a design is that the brightness loss in the monochromator due to aberrations or due to Coulomb interactions is minimal. Even when we the start with the very high brightness of the prototype 'high-brightness' gun used in our experiments, we do not measure noticeable loss of brightness in the monochromator except for the inevitable loss due to the removal of part of the energy spectrum. However, a complication of this monochromator design is that the beam at the exit of the monochromator is not free of energy dispersion and that special care has to be paid to this dispersion when the illumination is set. We demonstrate that when the dispersed source is properly focused on the specimen, the illumination can satisfy all requirements needed for high-brightness monochromated TEM illumination.

In an companion paper [18] we apply this monochromated illumination to focal series reconstructions and demonstrate information transfer down to 0.5 Å in the reconstructed exit waves.

2. Contrast transfer theory and information limit

2.1. Contrast transfer function

In the image plane, contrast is formed by interference between the undiffracted beam and the diffracted beams, and by interference between two diffracted beams. These two processes give linear and non-linear imaging, respectively. Usually, the contribution of non-linear imaging can be neglected compared to that of linear imaging, since the amplitudes of the diffracted beams are much smaller than that of the undiffracted beam. The linear contribution to the image intensity $I(G)$ is described by the well-known framework of linear imaging of weak phase objects (see Ref. [25]) as:

$$I(G) = \delta(G) - 2 \times \phi_p(G) \times \sin[2\pi\chi(G)] \times E_D(G) \times E_S(G) \times E_{vibr}(G) \quad (1)$$

where G is the spatial frequency, $\delta(G)$ is the Dirac delta function describing the contribution of the undiffracted beam, $\phi_p(G)$ is the phase shift induced by the specimen, $\sin[2\pi\chi(G)] \cdot E_D(G) \cdot E_S(G) \cdot E_{vibr}(G)$ is the partially coherent Contrast Transfer Function (CTF), and $\chi(G)$ is the phase aberration function:

$$\chi(G) = 1/2\lambda G^2 F + 1/4\lambda^3 G^4 C_S + 1/6\lambda^5 G^6 C_5 \ldots \quad (2)$$

(with wavelength λ, defocus F, spherical aberration C_S, and fifth-order spherical aberration C_5).

The intensity is reduced at high spatial frequencies by several effects. The effect of chromatic aberration is described by the temporal damping envelope

$$E_D(G) = \exp[-(\pi\lambda G^2 \Delta f)^2/2] \quad (3)$$

where Δf is the RMS of the focal spread given by:

$$\Delta f = C_C[(\Delta E/eV)^2 + (\Delta V/V)^2 + (2\Delta I/I)^2]^{1/2}, \quad (4)$$

C_C is the total chromatic aberration, ΔE/eV is the relative RMS energy spread of the electron gun, $\Delta V/V$ is the relative RMS instability of the high tension, and $\Delta I/I$ is the relative RMS instability of the objective lens current.

The effect of coherence angle, defocus and spherical aberration is described by the spatial coherence damping envelope:

$$E_S(G) = \exp[-\pi^2\alpha^2 G^2 (F + \lambda^2 G^2 C_S + \lambda^4 G^4 C_5)^2] \quad (5)$$

which assumes a Gaussian distribution of the coherence angles with RMS value α. Note that the RMS value of this two-dimensional Gaussian is equal to its 1/e-value. If the coherence angles are evenly distributed over an aperture disk of radius φ, then the spatial damping envelope is given by:

$$E_S(G) = J_1[\sqrt{2}\, 2\pi\alpha(GF + \lambda^2 G^2 C_S + \lambda^4 G^4 C_5)]/[\pi\sqrt{2}\alpha(GF + \lambda^2 G^2 C_S + \lambda^4 G^4 C_5)] \quad (6)$$

where J_1 is the Bessel function of the first kind of order one, and $\alpha = \varphi/\sqrt{2}$ is again the RMS value of the distribution of coherence angles.

The effect of specimen vibrations, stage vibrations, and deflector noise is described by the vibration damping envelope [26]:

$$E_{vibr}(G) = \exp[-2(\pi\Delta x G)^2] \quad (7)$$

where Δx is the RMS value of the apparent image displacements.