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A quantitative method for measuring small residual beam tilts in high-resolution transmission electron microscopy

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ABSTRACT

In a transmission electron microscope, electron illumination beam tilt, or the degree of deviation of electron beam from its optical axis, is an important parameter that has a significant impact on image contrast and image interpretation. Although a large beam tilt can easily be noticed and corrected by the standard alignment procedure, a small residual beam tilt is difficult to measure and, therefore, difficult to account for quantitatively. Here we report a quantitative method for measuring small residual beam tilts, including its theoretical schemes, numerical simulation testing and experimental verification. Being independent of specimen thickness and taking specimen drifts into account in measurement, the proposed method is supplementary to the existing “rotation center” and “coma-free” alignment procedures. It is shown that this method can achieve a rather good accuracy of 94% in measuring small residual beam tilts of about 0.1° or less.

1. Introduction

Recent decades have seen great improvements in resolution—down to sub-angstrom—in high-resolution transmission electron microscopy (HRTEM), owing to implementation of aberration correctors and monochromators in the microscopes [1–5]. This advance has made it possible to relate the structures of various materials to images in a quantitative way [6–11]. Nevertheless, more accurate image analysis puts forward higher requirements for determination of imaging parameters [9]. Among important imaging and diffraction parameters in TEM, a small residual electron illumination beam tilt, i.e., deviation from the optical axis of the microscope, is almost inevitable to occur even after careful alignment during operation. Thus far this has been a difficult parameter to measure accurately.

For aberration-uncorrected TEM, the influence of a beam tilt on image contrast of a crystal is more severe than that of a crystal tilt of the same magnitude, as previously demonstrated by Smith et al. [12]. It has also been known that when a beam tilt occurs in an uncorrected TEM, higher order aberrations may appear as lower order aberrations and can considerably influence imaging conditions [13]. For aberration-corrected TEM, a small beam tilt will not noticeably affect imaging conditions and can even be used as an advantage to compensate for small local crystal tilt [14], i.e., a beam tilt becomes equivellent to a crystal tilt in this case. Even though, it is always best to know the accurate beam tilt values for post-factum quantitative image contrast analysis, since more and more accurate image simulations are being used to quantitatively determine 2D [6,15,16] and 3D structures [8,9], and even to determine the atomic-scale compositions of materials [17]. In other applications, such as aberration and defocus spread measurement and tilt series wavefunction reconstruction, beam tilt is assumed to be a known parameter [13,18–20]. In such cases, knowing accurate residual beam tilt is a plus to achieve accurate interpretation of the data.

Numerical approaches for measuring and correcting beam tilts exist [21–32], and some of their key principles have been applied successfully as the standard alignment procedures in the modern HRTEM instruments, including “rotation center” alignment and “coma-free alignment”. As shown by Typke [22–26], Koster [27–30] and other researchers [31,32], the beam tilt and associated aberrations can be related by beam-tilt-induced image displacement (BTIID) in real space. By observing and measuring the BTIIDs, the related beam tilt can be estimated and therefore corrected. The rotation center method is sufficient for low- and medium-resolution work [28], whereas the coma-free method can achieve higher precision. The coma-free method [27,28] uses three images taken with 3 beam tilts ($-\boldsymbol{t}$, 0, $\boldsymbol{t}$), where $\boldsymbol{t}$ is an induced beam tilt. By comparing the image displacements of image ($\boldsymbol{t}$) and image ($-\boldsymbol{t}$) with respect to image ($\boldsymbol{0}$), the beam misalignment can be estimated and corrected. However, both the coma-free and rotation

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center methods are based on the weak-phase-object approximation, i.e., a thin crystalline or amorphous sample is preferred for measurement. In addition, for both the two alignment procedures, it is assumed that the BTIIDs are much larger than the specimen drift induced image displacements (SDIIDs) and therefore the later effects are neglected without taking into account. When the beam tilt becomes very small (e.g., less than 0.1°), the BTIIDs can be smaller than the SDIIDs that almost always exist in a microscope. For these reasons it is not always assured that the beam tilt can be diminished completely after the two alignments and small residual beam tilts may often exist. Hence, a supplementary method that can incorporate the effects of thick samples and specimen drifts is needed for accurately measuring the possible small residual beam tilts in HRTEM, as its applications become more and more quantitative.

In the present study, to accurately measure small residual beam tilts in HRTEM, we propose a method supplementing to the existing methods. The proposed method uses a data set of through-focus (TF) series of HRTEM images taken from a crystalline sample without limitation on its thickness, and takes into account the effect of specimen drifts. Firstly, it is shown that within the theoretical scheme of this method, in which a proper aperture is employed at the back-focal plane of the objective lens when recording the TF-series of images, for particularly selected reflections in the diffractograms (Fourier transform) of the HRTEM images, a rigorous linear relations exist without any approximation between their phases of the complex functions and the beam tilt. As such, the beam tilt values can be estimated from these linear relations. Secondly, it is demonstrated that for small tilts the BTIIDs are negligibly small as compared with the SDIIDs in this procedure, and therefore specimen drifts must be corrected in advance to obtain the beam tilt values. Fortunately, the errors in the phases introduced by correction of the SDIIDs are random errors and counteract each other, whereas the phase increments of the selected reflections are systematically accumulative as the defocus changes. Hence, the attempted linear relations for measuring the beam tilt can be revealed. At last, using the experimental images in a self-validated beam tilt experiment, it is demonstrated that small beam tilts can be measured with rather good accuracies by employing the proposed method.

2. Theory and simulation

2.1. Theoretical scheme

The transmission cross-correlation coefficient (TCC) theory on image formation in HRTEM, which considers not only the interference between the central beam and the diffracted beams, but also the interference among diffracted beams [14,33,34], is an accurate theory for describing partially coherent imaging. In the TCC theory, the diffractogram or Fourier transform, $I(\boldsymbol{K})$, of a HRTEM image intensity, $i(\boldsymbol{R})$, where $\boldsymbol{R}$ and $\boldsymbol{K}$ are the two-dimension spatial vectors in real space and in reciprocal space, respectively, can be expressed as follows:

$$I(\boldsymbol{K}) = \int_{\boldsymbol{K}'} \Psi(\boldsymbol{K})\Psi^*(\boldsymbol{K}'-\boldsymbol{K})T(\boldsymbol{K}',\boldsymbol{K}'-\boldsymbol{K})d^2\boldsymbol{K}' \tag{1}$$

where $\Psi(\boldsymbol{K})$ and $\Psi^*(\boldsymbol{K})$ are the exit wave function and its conjugate in reciprocal space, respectively, and $T(\boldsymbol{K}', \boldsymbol{K}'\text{-}\boldsymbol{K})$ is the so-called transmission cross coefficient (TCC).

When the illumination beam is not exactly parallel to the optical axis, but has a tilt, represented by a vector $\boldsymbol{t}$ in reciprocal space, the TCC theory expression for diffractogram $I(\boldsymbol{K})$ can be modified as follows [12,35]:

$$I(\boldsymbol{K}) = \int \Psi(\boldsymbol{K}'+\boldsymbol{t})\cdot\Psi^*(\boldsymbol{K}'-\boldsymbol{K}+\boldsymbol{t})T(\boldsymbol{K}'+\boldsymbol{t},\boldsymbol{K}'-\boldsymbol{K}+\boldsymbol{t})d^2\boldsymbol{K}' \tag{2}$$

with the modified TCC

$$\begin{aligned} &T(\boldsymbol{K}'+\boldsymbol{t},\boldsymbol{K}'-\boldsymbol{K}+\boldsymbol{t}) \\ &\quad = P(\boldsymbol{K}'+\boldsymbol{t})P^*(\boldsymbol{K}'-\boldsymbol{K}+\boldsymbol{t})E_\alpha(\boldsymbol{K}'+\boldsymbol{t},\boldsymbol{K}'-\boldsymbol{K}+\boldsymbol{t})E_\delta \\ &\qquad \times(\boldsymbol{K}'+\boldsymbol{t},\boldsymbol{K}'-\boldsymbol{K}+\boldsymbol{t}) \end{aligned} \tag{3}$$

$P(\boldsymbol{K})$ and $P^*(\boldsymbol{K})$ are the phase transfer function due to the lens aberrations and its conjugate, defined as $P(\boldsymbol{K})=\exp[-i\chi(\boldsymbol{K})]$, where $\chi(\boldsymbol{K})$ is the aberration function of the objective lens given by

$$\chi(\boldsymbol{K}) = 2\pi\left(\frac{1}{2}\lambda\cdot\Delta f\cdot\boldsymbol{K}^2 + \frac{1}{4}\lambda^3\cdot Cs\cdot\boldsymbol{K}^4\right) \tag{4}$$

$$E_\delta\left(\boldsymbol{K}',\boldsymbol{K}'-\boldsymbol{K}\right) = \exp\left\{-0.5(\pi\lambda\delta)^2\left[\boldsymbol{K}'^2-\left(\boldsymbol{K}'-\boldsymbol{K}\right)^2\right]\right\} \tag{5}$$

$$\begin{aligned} E_\alpha\left(\boldsymbol{K}',\boldsymbol{K}'-\boldsymbol{K}\right) = \exp\Bigg\{-\left(\frac{\pi\alpha}{\lambda}\right)^2\Big[&Cs\lambda^3\boldsymbol{K}'^3+\Delta f\lambda\boldsymbol{K}' \\ &-Cs\lambda^3\left(\boldsymbol{K}'-\boldsymbol{K}\right)^2-\Delta f\lambda\left(\boldsymbol{K}'-\boldsymbol{K}\right)\Big]^2\Bigg\} \end{aligned} \tag{6}$$

where Δf is the defocus, Cs is the third order spherical aberration, and λ is the wave length of the incident electron wave. $E_\alpha(\boldsymbol{K})$ and $E_\delta(\boldsymbol{K})$ are the damping envelope functions due to spatial and temporal partial coherence, respectively. Here δ is the focal spread distribution, α is the half angle of beam convergence [14]. It is noticed from Eqs. (5~6) that $E_\alpha(\boldsymbol{K}', \boldsymbol{K}'\text{-}\boldsymbol{K})$ and $E_\delta(\boldsymbol{K}', \boldsymbol{K}'\text{-}\boldsymbol{K})$ are real numbers and can be ignored when considering the phase terms below.

As seen in Eq. (2), generally there is no straightforward analytical relation between the diffractogram and the imaging parameters because of the integration operation. Therefore, removing the integration operation in Eq. (2) is the key to find an analytical relation between beam tilt and diffractogram. This can be realized for certain reflections by using an appropriate objective aperture for HRTEM imaging. In such an aperture-limited experimental setup, the information limit of the diffractogram is set to double that of the aperture-limited diffraction pattern, as shown schematically in Fig. 1a and b. In this case, the double-K diffractogram spots can be expressed as follows (using $2\boldsymbol{K}$ for clarity to describe an outmost double-K spot in the diffractogram, where $\boldsymbol{K}$ represents a reflection spot within the aperture.):

$$I(\boldsymbol{2K}) = \int_{\boldsymbol{K}'} \Psi(\boldsymbol{K}'+\boldsymbol{t})\Psi^*(\boldsymbol{K}'-2\boldsymbol{K}+\boldsymbol{t})T(\boldsymbol{K}'+\boldsymbol{t},\boldsymbol{K}'-2\boldsymbol{K}+\boldsymbol{t})d^2\boldsymbol{K}' \tag{7}$$

Assuming $\boldsymbol{t}$ is a tilt much smaller than a $\boldsymbol{K}$ in magnitude, the integrand function has contributions only when $\boldsymbol{K}'=\{0,\pm\boldsymbol{K}\}$, because of the aperture-limiting setup, i.e., for $\boldsymbol{K}'>\boldsymbol{K}$, $\Psi(\boldsymbol{K}')=0$. We further have

$$\begin{aligned} I(\boldsymbol{2K}) &= \Psi(\boldsymbol{t})\Psi^*(-2\boldsymbol{K}+\boldsymbol{t})T(\boldsymbol{t},-2\boldsymbol{K}+\boldsymbol{t}) \\ &\quad+\Psi(-\boldsymbol{K}+\boldsymbol{t})\Psi^*(-3\boldsymbol{K}+\boldsymbol{t})T(-\boldsymbol{K}+\boldsymbol{t},-3\boldsymbol{K}+\boldsymbol{t}) \\ &\quad+\Psi(\boldsymbol{K}+\boldsymbol{t})\Psi^*(-\boldsymbol{K}+\boldsymbol{t})T(\boldsymbol{K}+\boldsymbol{t},-\boldsymbol{K}+\boldsymbol{t}) \\ &= \Psi(\boldsymbol{K}+\boldsymbol{t})\Psi^*(-\boldsymbol{K}+\boldsymbol{t})T(\boldsymbol{K}+\boldsymbol{t},-\boldsymbol{K}+\boldsymbol{t}) \end{aligned} \tag{8}$$

since also $\Psi^*(-2\boldsymbol{K}+\boldsymbol{t})=0$, $\Psi^*(-3\boldsymbol{K}+\boldsymbol{t})=0$ due to the aperture-limited setup.

Therefore, for such double-K diffractogram spots Eq. (2) simplifies as follows.

$$I(\boldsymbol{2K}) = \Psi(\boldsymbol{K}+\boldsymbol{t})\Psi^*(-\boldsymbol{K}+\boldsymbol{t})T(\boldsymbol{K}+\boldsymbol{t},-\boldsymbol{K}+\boldsymbol{t}). \tag{9}$$

The phase function $p(2\boldsymbol{K})$ in these double-K diffractogram spots introduced by objective lens aberrations in Eq. (3) can be simplified as below.