The Error of Aberration Measurements in HRTEM Using Zemlin Tableaus

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In modern high-resolution transmission electron microscopy lens aberrations, in particular the strong third-order spherical aberration of the objective lens, severely limit the conditions for a directly interpretable imaging of object structures. In recent years hardware aberration correction [1] and software aberration correction of reconstructed exit wave functions [2, 3] have become feasible, thus fully exploiting the information limit of an instrument. Both methods require an accurate aberration measurement which is used to align the lens corrector [1, 4] or to correct an experimental exit wave function numerically [2, 3].

A number of instruments have approached information limits of 0.1 nm, or below, and next-generation transmission electron microscopes will advance to resolutions where already fifth-order aberrations play a role, requiring the measurement of 25 independent aberration coefficients.

A frequently used method for aberration measurement is the recording of a Zemlin tableau of diffractograms [5] taken from a thin amorphous object. The first-order aberrations defocus and two-fold astigmatism are measured from a diffractogram with the illumination untilted. Higher-order aberrations can be deduced from diffractograms with the illumination tilted systematically, because they induce aberrations, in particular an additional defocus and a two-fold astigmatism.

The measurement accuracy of the residual aberration function, comprising each of the aberrations, depends on the choice of illumination tilts \( t \), and in particular on the accuracy of the defocus and astigmatism measurement, \( D \), from single diffractograms. In this work we investigate the relation of defocus and astigmatism accuracy, \( D \), and the illumination tilt magnitude \( t \), for a given information limit \( g_{\text{max}} \), the inverse electron wavelength \( K \), and a given criterion to assess the residual aberration function.

The aberration analysis displayed in Table 1 shows: fifth-order aberrations induce up to a four-fold change of the induced defocus and two-fold astigmatism with respect to the tilt azimuth \( f \). The mean induced defocus and astigmatism depends on three different powers of \( t \); contributions with higher symmetry depend only on one or two different powers of \( t \). A proper choice of tilt magnitudes and azimuths seeks to construct an accurate aberration measurement by decoupling the different dependencies on \( t \) and \( f \). A reasonable choice is then: one diffractogram with the illumination untilted, an inner ring of 8 diffractograms with tilt \( t_1 \) at equal azimuthal spacing, and an outer ring of 16 diffractograms with a tilt \( t_2 \) larger than \( t_1 \) at equal azimuthal spacing. Hence a measurement of 75 independent induced aberrations, defocus and the two components of two-fold astigmatism, enters the least-squares fit to the desired 25 lens aberration coefficients. This Zemlin tableau represents one of the many choices balancing the number of diffractograms, in the light of recording time, with an excess of independent measurements to increase the measurement accuracy.

Next, an optimum value for the relative tilt magnitudes \( t_1 \) and \( t_2 \) was determined analytically by solving the Gaußian normal equations of the fit problem. If only defocus, \( c_{10} \), and the spherical aberrations of the third order, \( c_{30} \), and the fifth order, \( c_{50} \), are considered, then \( t_1 = 0.7 \ t_2 \) is a near-optimal choice. Considering all aberrations up to the fifth order this value still holds true, as was found by a numerical analysis of the Gaußian normal equations. Figure 1 (right) displays a plot of the defocus and astigmatism accuracy \( D \), versus \( t_1/t_2 \), needed to keep the residual aberration function within bounds for a future 200 kV instrument with an information limit of 0.08 nm. The bounds were set by Maréchal’s \( L/14 \) criterion [6]: then the r.m.s. deviation of the residual aberration...
function does not exceed $p/7$. The least stringent defocus and astigmatism accuracy for the particular set of parameters is 0.54 nm.

Finally, the dependence of the defocus and astigmatism accuracy $D$ on the tilt magnitude $t_2$ was investigated, at a relative magnitude of $t_1 = 0.7 \ t_2$. Increasing the tilt magnitude will generally increase the level of induced aberrations, which can be deduced from the aberration analysis displayed in Table 1. Therefore, with a given defocus and astigmatism accuracy $D$ the measurement error of the residual aberration function can be reduced by larger tilts. In other words, for larger tilts the demand on $D$ becomes less stringent. Figure 1 (left) displays $D$ needed to fulfil Maréchal’s criterion for a given tilt magnitude; for very large tilts $D$ approaches a level of $0.328 \ K / g_{\text{max}}^2$, given by residual defocus and two-fold astigmatism alone.

References

TABLE 1. Influence of single aberrations $c_{nk}$ on tilt-induced defocus and two-fold astigmatism; $n$ denotes order, $k$ symmetry. The first column indicates the symmetry of the induced aberration with respect to the tilt azimuth $f$; the first row indicates the dependence on the tilt magnitude $t$.

<table>
<thead>
<tr>
<th>Induced defocus</th>
<th>Induced astigmatism</th>
</tr>
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<tbody>
<tr>
<td>$1$ $t$ $t^2$ $t^3$ $t^4$</td>
<td>$1$ $t$ $t^2$ $t^3$ $t^4$</td>
</tr>
<tr>
<td>$0$ $c_{10}$ $c_{30}$ $c_{50}$</td>
<td>$c_{12}$ $c_{32}$ $c_{52}$</td>
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<tr>
<td>$2$ $c_{21}$ $c_{41}$</td>
<td>$c_{21}, c_{23}$ $c_{41}, c_{43}$</td>
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<td>$3$ $c_{32}$ $c_{52}$</td>
<td>$c_{30}, c_{34}$ $c_{50}, c_{54}$</td>
</tr>
<tr>
<td>$4$ $c_{43}$</td>
<td>$c_{41}, c_{45}$</td>
</tr>
<tr>
<td>$c_{54}$</td>
<td>$c_{52}, c_{56}$</td>
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</tbody>
</table>

Defocus and astigmatism accuracy $D$ [nm]

![Graph showing the dependency of the r.m.s. defocus and astigmatism accuracy $D$ on the outer illumination tilt $t_2$. Inner tilt $t_1 = 0.7 \ t_2$, $K = 400$ nm$^{-1}$; upper: $g_{\text{max}} = 8$ nm$^{-1}$; lower: $g_{\text{max}} = 12$ nm$^{-1}$.](image1)

Relative magnitude of beam tilts $t_1/t_2$

![Graph showing the dependency of the r.m.s. defocus and astigmatism accuracy $D$ on the relative magnitude of inner and outer illumination tilt. Outer tilt $t_2 = 10$ nm$^{-1}$, $K = 400$ nm$^{-1}$, $g_{\text{max}} = 12$ nm$^{-1}$.](image2)