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Aberration corrected TEM: current status and future prospects

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Abstract. Aberration correction leads to a substantial improvement in the interpretable resolution of Transmission Electron Microscopes. Electron optical correctors based on two strong hexapole elements linked through a round lens transfer doublet enables direct correction of all axial aberrations to third order. Subsequent, indirect computational analysis of a focal or tilt series of images offers the possibility of further compensation of the axial aberrations to fifth order. This paper describes 1st and 2nd generation aberration corrected instrumentation installed in Oxford and also the use of combinations of direct and indirect correction / compensation in a variety of different geometries to achieve specimen exit plane wavefunctions containing directly interpretable structural information significantly below 0.1nm.

1. Introduction
In the past decade there has been a rapid development of electron optical components for the correction of spherical aberration in the Transmission Electron Microscope (TEM) and this has led to a dramatic improvement in instrumental resolution and spectroscopic capability. These developments as first reported at the EMAG conference held in Cambridge in 1997 and subsequently published elsewhere [1] have been made possible due to the development of complex electron optical components for the correction of the intrinsic spherical aberration always present in conventional electromagnetic round lenses [2]. The correcting elements also enable the correction of residual parasitic aberrations to third order using current electron optical geometries. Aberration corrected TEM instruments (of which there are in excess of 50 commercial instruments installed worldwide with either single, or more rarely double corrector geometries) have enabled the acquisition of atomic resolution data from a wide range of materials and to determine electronic structure and composition with single atom sensitivity (see [3] for a review).

Improvements beyond those that can be achieved using direct aberration correction can be made by utilising aberration corrected image data to recover the specimen exit plane wavefunction in one of several possible indirect schemes (see [4] for a review). In these methods the higher order aberrations can be locally measured and computationally compensated a posteriori to further extend the interpretable resolution. In this article, we review the current status of corrected instrumentation and outline the above two approaches to aberration correction emphasising the additional benefits of applying them in combination. Finally, prospects for the direct correction of higher order aberrations and the possibility of correction of chromatic aberration will be discussed.
2. Optics and instrumentation

The majority of aberration corrected TEMs incorporate correctors based on a design originally due to Rose and Haider [1] in which the primary correcting elements consist of a two strong hexapoles coupled through a round lens doublet. Correction is achieved due to the fact that the primary second-order aberrations of the first hexapole (a strong three fold astigmatism) are exactly compensated by the second hexapole element. Owing to their non-linear diffraction power, the two hexapoles additionally induce a residual secondary, third-order spherical aberration which is rotationally symmetric with a negative sign, thus cancelling the positive spherical aberration of the objective lens.

One of the first instruments using this corrector design (a Jeol 200kV 2200FS) for both probe and imaging aberration correctors and an in column energy filter was installed in Oxford in 2003 (Figure 1(a)) [5]. Adjustment of the imaging corrector is achieved using a Zemlin tableau of diffractograms calculated from images of a thin amorphous foil and recorded at several tilt azimuths with constant tilt magnitude [6]. These datasets provide measurements of the tilt-induced defocus and 2-fold astigmatism yielding linear estimates for the coefficients of the wave aberration function. In practice, a computationally efficient algorithm is used in which experimental diffractograms are compared to a library of pre-calculated diffractograms such that iterations of the tuning procedure can be completed in less than 30 s.

More recent instruments (see for example Figure 1(b)) feature additional stabilisation of the microscope column to reduce mechanical vibration in addition to improved stabilities in both the high voltage and objective lens supplies to values of typically $ca. 5 \times 10^{-7}$. A number of installations also feature monochromated electron sources that enable a reduction in the beam energy spread to $ca. 0.1$ eV reducing the effects due to temporal coherence and offering spectroscopic energy resolution directly comparable with that achievable from cold field emission sources. At the time of writing,
these second generation instruments are capable of delivering directly interpretable TEM resolutions of less than 80 pm at primary voltages between 200 and 300 kV (Figure 2), limited only by the effects of chromatic aberration and residual mechanical, acoustic and electrical instabilities. Future developments will address these through the incorporation of further mechanical and acoustic isolation and more complex corrector geometries that enable the higher order coefficients in the wave aberration functions and the correction of chromatic aberration [7].

Figure 2 Youngs fringe diffractogram calculated from images of a gold particles supported on an amorphous C film using the Oxford Mark 2 aberration corrected instrument (Jeol 2200MCO) operated at 200kV. Clear information transfer extending to 0.07 nm is visible with addition reflections (marked) from the gold particles visible at 0.05 nm.

3. Optimum corrected imaging conditions

Optimum phase contrast imaging conditions for conventional high resolution TEM are set by balancing the phase shifts due to the fixed, positive third-order spherical aberration ($C_3$) with an appropriate compensating negative defocus ($C_1$) according to the well known Scherzer condition [2] given by:

$$C_1 = -\frac{\lambda^{3/2}}{C_3^{1/2}}$$

which leads to an interpretable resolution limit of

$$d = 0.625\lambda^{3/4}C_3^{1/4}$$

For aberration corrected TEMs two alternative conditions can be defined. The first balances the positive $5^{th}$ order spherical aberration ($C_5$) against both the third order spherical aberration and defocus (noting that this leads to an optimal negative $C_3$ and positive $C_1$) [2,8] as

$$C_3 = -2.88\lambda^{3/2}C_5^{2/3}$$
$$C_1 = 1.56\lambda^{2/3}C_5^{1/3}$$
In turn this defines an interpretable resolution limited by $C_5$ given by

$$d = 0.625 \lambda^{5/6} C_5^{1/6} \quad (4)$$

An alternative optimal condition has also been proposed in where the chromatic aberration ($C_c$) is limiting [8]. In this case alternative aberration settings can be defined to ensure that the first zero in the phase contrast transfer function coincides with the information limit due to temporal coherence. Under these conditions the appropriate values for $C_1$, $C_3$ and $C_5$ are given by

$$C_1 = 1.7 \pi \Delta$$
$$C_3 = -3.4 \left( \frac{\pi \Delta}{\lambda} \right)^2$$
$$C_5 = 1.3 \left( \frac{\pi \Delta}{\lambda^2} \right)^3 \quad (5)$$

with

$$\Delta = C_c \left\{ \left( \frac{\Delta V}{V} \right)^2 + 4 \left( \frac{\Delta I}{I} \right)^2 + \left( \frac{\Delta E}{V} \right)^2 \right\}^{1/2} \quad (6)$$

The above define suitable operating conditions for optimal phase contrast imaging. However both $C_3$ and $C_1$ may be set to zero leading to pure amplitude contrast [9]. Under this latter condition the phase contrast transfer function has a value of $0$ while the amplitude-contrast transfer function is maximised. Importantly, this imaging mode is not available in standard uncorrected TEMs.

### 4. Indirect compensation and exit wave reconstruction

Several methods have previously been developed for the experimental determination of the axial aberration coefficients (see [4] for a review) all of which are based on the injection of known multiple beam tilt geometries and the measurement of the induced first order aberrations. These provide estimates of the coefficients of the wave aberration function, $W(\theta, \phi)$, expanded here to third order and expressed in polar form to make the radial and azimuthal dependence of the coefficients apparent. Such measurements can subsequently be used in the recovery of both the phase and modulus of the specimen exit-plane wavefunction [4] under either linear or non-linear imaging models.

$$W(\theta, \phi) = | A_0 | \theta \cos(\phi - \phi_1)$$
$$+ \frac{1}{4} | A_1 | \theta^2 \cos 2(\phi - \phi_{22}) + \frac{1}{2} C_1 \theta^2$$
$$+ \frac{1}{4} | A_2 | \theta^3 \cos 3(\phi - \phi_{33}) + \frac{1}{4} | B_2 | \theta^3 \cos(\phi - \phi_3)$$
$$+ \frac{1}{4} | A_3 | \theta^4 \cos 3(\phi - \phi_{44}) + \frac{1}{4} | S_3 | \theta^4 \cos 2(\phi - \phi_{32}) + \frac{1}{4} C_3 \theta^4 + \ldots \quad (7)$$

This approach has demonstrated resolution enhancement for uncorrected microscopes and more importantly also complements direct aberration correction.

Direct correction has the advantage that it may be achieved on line using a single image and does not require post acquisition processing. However with current generation optical elements, correction of aberrations in the TEM extends to third order only and the recorded data comprises intensity only. In contrast, indirect compensation and restoration recovers the complex specimen exit plane wavefunction.
Wavefunction and compensation of the aberration coefficients to any order is theoretically possible, limited only by the measurement accuracy. The disadvantage is that this is an off-line technique requiring post acquisition processing and the recording of multiple image datasets with a consequent increase in radiation exposure.

Indirect and direct methods can also be used in combination [10] and provide further advantages. For the focal series geometry, the elimination of tilt-induced axial coma relaxes the requirement of using parallel illumination. Thus, current density at the sample may be maintained while reducing the emitter current and converging the illumination thereby giving a reduced energy spread and hence improved information limit. For a tilt series dataset, the elimination of tilt induced axial coma gives rise to less critical focus conditioning for a given tilt magnitude [10] and multiple tilt magnitudes are possible without any induced focus change. Localised compensation of higher order aberrations up to 5th order is also possible.

Figure 3 illustrates the cumulative effects of initial direct electron optical correction followed by subsequent aberration refinement on data obtained from a nanocrystalline Pt catalyst particle [11], demonstrating the improvement obtained by recovering the specimen exit wavefunction compared to the information present in a single HRTEM image. The power spectra shown below each image in Figure 3 show successively the resolution enhancement obtained from aberration correction and the presence of higher spatial resolution information in the reconstructed specimen exit plane wavefunction together with a decreased contribution to the power spectrum from the amorphous carbon support.

Figure 3 (a, b) HRTEM (intensity) images of a nanocrystalline Pt catalyst particle acquired with Cs uncorrected at 0.5 mm (a) and corrected to -30 μm (b). Corresponding power spectra are shown beneath each image. (c) Phase of the specimen exit plane wavefunction and power spectrum of the same particle, obtained after aberration compensation and refinement of a through-focus series of aberration-corrected images.
5. Conclusions and future prospects

This short paper has summarised the current status of aberration correction in the TEM with particular reference to instrumentation and corrector optics. The current generation of intermediate voltage corrected TEMs are capable of full correction of all aberration coefficients to third order (and by adjustment of the beam trajectories within the corrector, partial compensation of selected 4th and 5th order coefficients). These instruments are now providing structural data at resolutions exceeding 80 pm, limited largely by the effects of temporal coherence.

Future instruments are likely to address this through the incorporation of complex second generation correcting elements with or the correction of all axial aberrations to fifth order in addition to chromatic correction. If these developments are successful and sufficient instrumental and environmental stability can be achieved then it is likely that TEM resolution will be further increased to levels approaching 50 pm.

Concurrently we can also expect to see advances in in-situ microscopy where aberration correction enables the objective lens polepiece gap to be enlarged without significant increases in C₃ and a consequent deterioration in resolution, thus permitting more complex sample treatment and manipulation facilities to be fitted.

Finally we note that aberration correction in the TEM will also impact imaging of radiation sensitive biological materials at higher resolution than is currently possible. However, for these applications, to recover the lower spatial frequencies the combined use of imaging and aberration corrector coupled to a suitable electrostatic phase plate inserted in the back focal plane of the objective lens [12] may be optimal.

References

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