Aberration corrected tilt series restoration

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Abstract. Aberration correction can be achieved using either direct electron optical correction or indirect image restoration. In the past focal series restoration has been applied to aberration corrected images in order to improve the quality of aberration correction and retrieve the complex specimen exit wavefunction. Image restoration can also be performed from a number of images with differing illumination tilts (a tilt series) instead of the more common focal series datasets where only the defocus value is varied. Here we apply tilt series image restoration to aberration corrected images and discuss the advantages of this approach. Preliminary results demonstrate the potential of this technique to provide interpretable structural information at resolutions beyond the axial information limit of the microscope.

1. Introduction
The interpretation of conventional high resolution transmission electron microscope (HRTEM) images is complicated by the aberrations of the objective lens. These can be removed by either direct or indirect methods of aberration correction. The former involves the insertion of multipole elements in the electron optical column that correct the inherent positive spherical aberration [1]. Indirect methods require a dataset of multiple images with differing values of defocus or illumination tilt, from which the aberrations may be measured and computationally compensated \textit{a posteriori} to recover the specimen exit wavefunction (see [2] for a review). It is also possible to combine both direct and indirect approaches and to recover the specimen exit wavefunction from aberration corrected images [3].

In this paper, we discuss the advantages of applying indirect restoration procedures to aberration corrected images and present the results of the first application of tilt series restoration to aberration corrected images.

2. Combining Direct and Indirect Methods
Direct correction offers the advantage that it may be achieved on line with a single image and does not require post acquisition processing. However, at resolutions below 0.1nm the effects of higher order aberrations become increasingly significant and current generation optical elements are not able to correct aberrations above third order [4]. In contrast, using indirect methods correction to any order is theoretically possible, limited only by the measurement accuracy. A further advantage of the indirect approach is that it recovers the complex exit wave whereas the recorded data in a single corrected image.

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image comprises intensity only. This means that the highly sensitive phase information is lost. The disadvantage is that indirect correction is an off-line technique requiring multiple image datasets.

Further advantages are obtained when indirect and direct methods are used in combination. Direct electron optical correction of the aberrations up to 3rd order enables accurate measurement of higher order aberrations, facilitating a localised indirect compensation up to 5th order [5]. Post imaging indirect compensation also allows compensation of residual lower order aberrations. Figure 1 illustrates typical phase plates obtained after initial electron optical correction to third order, and after compensation to fifth order using locally determined values of the aberration coefficients, with the latter showing a clear improvement in the extent of transfer within a $\pi/4$ phase limit to sub 0.1nm levels.

![Figure 1](image1.png)

**Figure 1.** Phase plates calculated to 5th order from residual aberrations a) After direct electron optical correction to third order, b) After local refinement of aberrations to 5th order. Black to white represents a phase shift of $\pi$. Inner and outer circles represent resolution limits of 0.1 and 0.08nm.

Correction of the coherent aberrations up to 3rd order also greatly reduces tilt-induced axial coma, which 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. These benefits apply equally to both tilt or focal series restoration.

### 3. Advantages of tilt series restoration under aberration corrected conditions

Unlike for focal series datasets, the resolution of a tilt series restoration is not limited to the axial information limit of the microscope as determined by the spatial and temporal coherence. In a conventional uncorrected TEM the defocus condition for tilted imaging must be chosen very accurately in order to achieve reasonable information transfer. These practical challenges with data acquisition limit the tilt angle that can be used in the restoration for a given electron wavelength ($\lambda$) and spherical aberration ($C_s$) to a compromise value of $(\lambda/C_s)^{1/4}$ [6]. This tilt angle is typically a few mrad so severely limits the resolution improvement that is achievable. However, in a transmission electron microscope capable of direct electron optical aberration correction the value of axial coma is much less sensitive to the illumination tilt angle. This gives rise to less critical focus conditioning for a given tilt magnitude. Direct aberration correction also results in a smaller tilt induced defocus change. These two effects mean that under aberration corrected conditions the dataset for successful tilt series restoration is less dependant on optimal microscope stability and imaging conditions. This allows the use of larger tilt angles up to 18 mrad (c.f. optimal tilt angle of 3 mrad for an uncorrected microscope) so that greater resolution improvements are achievable.
Figure 2. Moduli of Fourier transforms taken from the complex specimen exit waves of \( \langle 111 \rangle \) orientated SrTiO\(_3\) produced using (a) a focal series data set and (b) a combined tilt / focal series data set. The 10% information limit for these restorations corresponds to 0.13 nm and 0.09 nm and the axial information limit is marked on both figures for reference. Selected \( \langle 033 \rangle \) type reflections corresponding to 0.092nm resolution are highlighted in tilt series data to emphasize the presence of higher resolution information compared to the exit wave recovered from the axial focal series data. Asymmetry in the transform in (b), for example the contrast difference between the \([3-30]\) and \([-330]\) reflections, indicates a very small crystal misalignment and this is the cause of the slightly square appearance of the atomic columns shown in the phase (c) and modulus (d) restored from the combined tilt / focal series data set.

4. Application of tilt series restoration for aberration corrected images

To test the feasibility of aberration corrected tilt series restoration we have initially studied materials with well characterised atomic structures. Images of SrTiO\(_3\) were recorded using a JEOL JEM-2200FS C\(_S\)-corrected instrument operated at 200kV with a spherical aberration of -3\( \mu \)m [7]. At the microscope, aberrations were measured to 5th order from diffractograms of amorphous carbon and electron optically corrected to 3rd order [8]. Images were energy filtered using a post-specimen omega-filter and pre-processed to correct for the effects of the modulation transfer function of the CCD camera.

In order to demonstrate that the information transfer of aberration corrected tilt series restoration is beyond the axial limit we have also compared this approach with the axial focal series approach.
Focal series comprised of 20 images separated by a focal increment of 8 nm with the series centered about the Gaussian focus condition. These conditions are optimal for focal series restoration of aberration corrected data. The tilt series restoration described in this work comprised 27 images [9]. This includes three axial images at defoci of 8nm, 24nm and 31nm overfocus, used to produce an initial estimate of the wavefunction. Similar short focal series are taken at six different tilt azimuths at a tilt angle of 16 mrad using intermediate axial images for registration. As the samples used were estimated to be less than 2 nm thick parallax effects could be ignored and a linear imaging approximation could be used during the restoration procedure.

For both datasets, specimen exit waves were restored under the linear imaging approximation using a Wiener restoring filter. The aberration measurements were then locally refined to 3rd order for a specimen sub-region of interest using the phase correlation function / phase contrast index approach to an accuracy of <1 nm [10]. Figure 2 demonstrates the significant improvement in resolution obtainable from an aberration corrected tilt series restoration compared to an equivalent aberration corrected focal series. For the focal series restoration the information transfer limit of 10% corresponds to a spatial resolution of 0.13 nm, whereas in the tilt series restoration it corresponds to 0.091 nm in all directions.

5. Conclusions
Focal series restoration applied to aberration corrected images improves the signal to noise ratio and retrieves the complex specimen exit wave but cannot increase the resolution beyond the axial information limit of the microscope. In contrast, it has been shown in this paper that aberration corrected tilt series restoration is able to achieve significant resolution improvement beyond the classical axial information limit.

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