Aberration-corrected STEM: current performance and future directions

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Abstract. Through the correction of spherical aberration in the scanning transmission electron microscope (STEM), the resolving of a 78 pm atomic column spacing has been demonstrated along with information transfer to 61 pm. The achievement of this resolution required careful control of microscope instabilities, parasitic aberrations and the compensation of uncorrected, higher order aberrations. Many of these issues are improved in a next generation STEM fitted with a new design of aberration corrector, and an initial result demonstrating aberration correction to a convergence semi-angle of 40 mrad is shown. The improved spatial resolution and beam convergence allowed for by such correction has implications for the way in which experiments are performed and how STEM data should be interpreted.

1. Introduction

There are now several scanning transmission electron microscope (STEM) instruments operating that have been fitted with spherical aberration correctors. Correction of spherical aberration is revolutionizing the performance of STEM instruments. Not only does it provide dramatically improved spatial resolution [1], but also improves by an order or magnitude or more the beam current at a given resolution [2], and allows resolving information in the third (specimen depth) dimension [3]. The HB603U instrument at Oak Ridge National Laboratory has been equipped with a Nion aberration corrector, and has recently clearly resolved an atomic column spacing of 78 pm, with transfer demonstrated to 61 pm [1]. In this paper we will describe the technological developments and experimental practices that were necessary to achieve this resolution. Many of the difficulties encountered in achieving this result will be overcome in a next generation instrument that has been developed by Nion and is currently under test, and here we illustrate its aberration-correcting capabilities and projected performance.

The ability to form electron probes at or below 50 pm in size provides opportunities and challenges. Many hitherto unfeasible experiments become possible, but there are challenges in interpreting the data resulting from these experiments.
2. Current performance of \( C_s \) corrected STEM instruments

As an example of an image recorded on an aberration-corrected STEM instrument, figure 1 shows an annular dark-field (ADF) image of Si oriented in the \(<112>\) direction recorded on a VG Microscopes HB603U instrument fitted with a Nion aberration corrector. The HB603U instrument has a 300 kV cold field-emission gun (CFEG), and its objective lens has a \( C_s \) of 1 mm. Prior to correction, this instrument had a point resolution limit in the ADF imaging mode of 0.13 nm (130 pm).

When oriented to \(<112>\) the atomic positions project into columns separated by 78 pm, and these columns are clearly resolved. The Fourier transform of the image (figure 2) shows a spot corresponding to the \( (444) \) planes corresponding to the 78 pm column separation. Furthermore, closer examination of figure 2 shows that there is visible transfer as far as the \( (804) \) planes with a spacing of 61 pm. The dramatic improvement of ADF STEM through correction of \( C_s \) is therefore obvious, and it is worth considering some of the technical developments that have allowed this improvement and the current limitations:

Figure 1. (a) Unprocessed STEM ADF image of Si\(<112>\). The dumbbell pairs of atomic columns are separated by 78 pm. (b) A line profile through the image as indicated, summing over a line 10 pixels wide.

Figure 2. A. A Fourier transform of the image in figure 1 with a modulus line profile through the region shown showing transfer to the 804 (61 pm) spacing.

2.1. Aberration measurement and parasitic aberration correction
To achieve a resolution of 61 pm at 300 kV accelerating voltage requires precise measurement and control of the aberrations. Any misalignment or inhomogeneity of the multipole magnetic fields used will lead to parasitic aberrations. Additional multipole windings must be provided to compensate for these parasitic aberrations, and the existing Nion corrector uses a total of 33 separate windings and power supplies. The current in these windings needs to be carefully adjusted to precisely null the parasitic aberrations. The precisions to which some of the aberrations must be measured to achieve 61 pm at 300 kV are shown in Table 1. Accurate and automated tuning software [2] based on Ronchigram analysis is used to provide an easy-to-use corrector tuning system that does not require detailed knowledge of aberrations.

Table 1: The measurement precision required for some example aberrations to achieve 61 pm imaging at 300 kV.

| Aberration                  | Defocus \((C_{1,0})\) | Coma \((C_{2,1})\) | Threefold astigmatism \((C_{2,3})\) | Spherical aberration \((C_{3,0})\) |
|-----------------------------|------------------------|---------------------|--------------------------------------|----------------------------------|
| Required precision          | 1.2 nm                 | 151 nm              | 92 nm                                | 12 \(\mu m\)                     |

2.2. Microscope stability
It is often said that aberration-correctors correct aberrations but not instabilities. Although the aberration corrector itself can be designed to be extremely mechanically and electrically stable, the HB603U column was designed many years ago when the achievement of 130 pm resolution was pushing the limits. Finding and correcting sources of instabilities in the HB603 column was a time-consuming job that was necessary to allow the improved spatial resolution. Such instabilities include both electrical and mechanical sources.

2.3. Higher order aberrations
Although the Nion corrector that is used on VG instruments is capable of correcting the third-order aberrations (such as \(C_3\)) its operation does generate significant 5th order aberrations. The explanation lies in the fact that the third order correction provided by the corrector octupoles does not occur in a plane that is optically conjugate to the objective lens. The propagation of the beam over the resulting optical length results in combination aberrations. After careful measurement of the 5th order aberrations, it is possible to partially compensate for their effect using non-zero 3rd and 1st order aberrations, in a similar way to the use of underfocus to compensate for uncorrected spherical aberration. The measured values of the 5th order aberrations, and the settings of the lower order aberrations to provide optimal compensation, are shown in Table 2. Although the value of the \(C_{5,0}\) aberration is similar to those measured in Nion corrected 100 kV machines and requires careful compensation, it is interesting to note that the \(C_{3,2}\) and \(C_{3,4}\) values are lower than those of the 100 kV machines due to the slightly different beam configuration. These small values actually result in compensating lower order aberrations that are smaller than the required measurement precision (see Table 1), and the compensation is therefore not so critical for these aberrations.

Table 2: The 5th order aberrations measured prior to acquiring the image in figure 1, and the settings of the 3rd and 1st order aberrations used to compensate

| Aberration | Measured value | 3rd order compensation | 1st order compensation |
|------------|----------------|-------------------------|------------------------|
| \(C_{5,0}\) | +100 mm        | \(C_{3,0}\)(spherical ab.) = -37 \(\mu m\) | \(C_{1,0}\)(defocus) = +1.9 nm |
| \(C_{5,2}\) | -27 mm         | \(C_{1,2}\) = +9.8 \(\mu m\) | \(C_{1,2}\)(astig.) = -0.5 nm |
| \(C_{5,4}\) | +22 mm         | \(C_{3,4}\) = -5.2 \(\mu m\) |                        |
| \(C_{5,6}\) | +1.5 mm        |                         |                        |
2.4. Chromatic aberration

The correction of chromatic aberrations requires opposing deflecting electrostatic and magnetic fields and is more difficult to achieve because of the high stabilities necessary when fields are opposed, and practical issues (such as insulator breakdown or shorts) involved in applying large electrostatic fields. For this reason $C_C$ correction has so far been developed commercially only for lower voltage scanning electron microscope instruments. For the HB603U instrument, the effect of $C_C$ (measured as 1.6 mm) is small for a spatial resolution of 61 pm because of the low energy spread of the CFEG compared to the primary beam energy.

Using the measured values of all the aberration values up to and including 5th order, and incorporating the effect of $C_C$ and finite electron source size results in the probe intensity profile shown in figure 3. A convolution of this probe with an object function consisting of Lorentzians with width 30 pm located at the atom sites leads to a simulated image that closely resembles the experimental image. A significant constant background was added to the simulated profile to achieve this match because the experimental peak-to-peak intensity variation in figure 3b is approximately 1/4 of the total detected signal. An amorphous scattering layer, and scattering by dechannelled electrons, may be responsible for this background.

3. A next generation STEM column and aberration corrector

Nion is currently in the testing phase of the development of an entirely new STEM column and aberration corrector designed to address many of the issues listed in section 2. It is a modular column, but in its as-designed configuration, the illumination system consists of 2 round condenser lenses followed by a 19 layer corrector, followed by a condenser-objective lens. The first-two columns of this design are being built on existing VG built 100 kV CFEGs, but the third will be fitted with an entirely new 200 kV CFEG also being designed by Nion.

The new column is designed to be highly mechanically, electrically and thermally stable in order to provide a suitable platform for the sub-50 pm resolution anticipated. The objective lens has been designed to have a very low $C_C$ coefficient of 0.8 mm. With the rest of the column providing only a further 0.3 mm, the total $C_C$ of the system is expected to be about 1.1 mm.

A next generation aberration corrector has been designed for the new column that is capable of correcting all aberrations up to and including 5th order. Calculations of the electron trajectories show that the limiting aberrations with be of 7th order with about 500 mm of both $C_{7,0}$ and $C_{7,2}$. Suitable compensation by the 5th, 3rd and 1st order aberrations would, in the absence of $C_C$, allow beam convergence semi-angles of up to 55 mrad to be used. Figure 3 shows a Ronchigram taken during
testing of the new column. Despite the complexity of the new corrector, it can be seen that the flat-phase region of the Ronchigram extends to 40 mrad, with further improvement expected following further optimisation of the electron trajectories within the corrector. In practice, the effects of $C_0$ will limit the usable maximum convergence angle to 40 mrad for 100 kV and 50 mrad at 200 kV. Taking into account other resolution limiting factors, we anticipate performance approaching 50 pm at 200 kV.

![Figure 3. A Ronchigram collected from the next generation Nion column fitted with a $C_0/C_3$ corrector showing correction to 40 mrad. The expected maximum angle of geometric aberration correction will be 55 mrad.](image)

4. Consequences of increased beam convergence
Correction of spherical aberration allows a larger aperture to be used in the objective lens, resulting in the improved spatial resolution and increased beam current. The converging beam that is collapsing to form the illuminating probe therefore has a larger convergence angle. The possibility of this convergence angle reaching to 50 mrad creates somewhat of a paradigm shift in the way we can interpret STEM images.

4.1. Validity of the local approximation
Annular dark-field imaging in the STEM can be regarded as being an incoherent imaging process. One explanation for this is that at the high scattering angles collected, the scattering is predominantly thermal diffuse. To simulate ADF images therefore requires careful consideration of both the total incoherent scattering, and that fraction that reaches the ADF detector. These are usually both treated in terms of an incoherent local scattering potential, $V'(r)$, that is multiplied by the wavefunction intensity within the crystal.

In reciprocal space, incoherent scattering can be incorporated through the use of a mixed dynamical form factor (MDFF) that describes the coupling between plane waves $g$ and $h$ into a final scattering vector, $s$. For thermal diffuse scattering making the Einstein phonon dispersion approximation, the MDFF can be written,
\[ \mu(s, g, h) = f_{el}(s - g) f_{el}(s - h) \sum \exp[-i(g - h) \cdot r_i] \times \left\{ \exp[-M(g - h)^2] - \exp[-M(s - g)^2] \exp[-M(s - h)^2] \right\} \]  

where \( M \) is the usual Debye-Waller factor and the sum is over atomic locations. To express a local incoherent scattering potential in real-space, the MDFF must reduce to being a function of \( g \cdot h \). If both \( g \) and \( h \) are small compared with the typical scattering angles, \( s \), at which TDS occurs, then eq. (1) can be approximated in this way. Aberration correction, however, leads to much larger \( g \) and \( h \) values for which it is not clear that the local approximation is valid. In real-space this argument can be equivalently expressed as asserting that the TDS scattering from neighbouring atoms will be incoherent due to their uncorrelated vibrations, but any given atom will still be coherent with respect to itself. Once the probe size becomes comparable to the atomic scattering potential, coherent effects may start to manifest themselves even in thermally scattered electrons.

4.2. Reduced depth of field and dechannelling

Annular dark-field imaging and high-spatial resolution EELS has long made use of the strong channelling conditions that occur when an atom-sized probe illuminates an atomic column in a crystal. This channelling occurs because the probe strongly couples into the bound s-eigenstates of the high-energy wavefunction. Calculations show that this coupling peaks at a probe convergence semi-angle of about 20 mrad at 300 kV [4]. Beyond this angle, the illuminating beams couple into higher angle states that are only weakly scattered and approximate to plane-wave states. These higher angle beams can therefore provide a reduced depth of focus of the illuminating beam in the crystal, and can be used to provide depth sensitivity for both imaging and spectroscopy [3]. Clearly, careful consideration now needs to be given to the degree to which images may be interpreted as either a projection or as an optical section of a thin region within the sample. Similarly, the effect of the increased beam spreading due to the non-channelling high-angle beams also needs to be taken into account for a fully quantitative analysis.

5. Conclusions

We have demonstrated the dramatic improvement in performance that occurs when the spherical aberration of a STEM instrument is corrected, and explained some of the experimental considerations necessary to achieve this performance. The next generation of instruments will perhaps achieve their optimal performance more routinely.

Aberration correction also dramatically changes the conditions under which we perform experiments, and it is time to re-evaluate the way we interpret the data. For example, rather than relying on channelling to provide a projection of the sample structure, we now need to consider dechannelling effects, but these can be used to provide depth sensitivity. In a similar vein, it is timely to re-evaluate the approximations commonly made in STEM image simulations in light of the new experimental conditions.

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