Towards sub-0.5 angstrom beams through aberration corrected STEM

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Abstract. Correction of spherical aberration (C_S) in the scanning transmission electron microscope (STEM) has enabled routine sub-angstrom resolution imaging and increased the current available in an atom-sized probe by a factor of 10 or more. Both high-angle annular dark field (HAADF) imaging and spectrum imaging (SI) results are shown from instruments fitted with Nion aberration correctors

1. Introduction

Improving the performance of an electron microscope by aberration correction has recently progressed from being an unattainable wish to a commercially available reality. A C_S corrected dedicated STEM operating at 120 kV has demonstrated sub-angstrom spatial resolution [1], and a corrected 300 kV STEM is routinely imaging at resolutions better than 0.8 Å [2]. In a high-resolution transmission electron microscope (HRTEM), the ability to adjust the phase contrast transfer function through control of C_S has allowed O atoms in an oxide material to be observed for the first time, with the machine operating at a spatial resolution of 1.3 Å [3].

The first theoretical design for a C_S corrector was proposed in 1947 [4], but it took more than half a century before practical aberration correctors were able to demonstrate an improvement in resolution over similar uncorrected machines. One of the reasons for this is that, contrary to popular perception, the lenses in electron microscopes are already of high optical quality making it hard to improve upon them. In a previous paper [5] we have shown how even an uncorrected high resolution microscope can be judged to be optically superior to the corrected Hubble space telescope. Indeed, a significant fraction of the C_S of a modern high resolution objective lens can be explained by geometry and deviations from the small angle approximation. Consider a so-called Gaussian lens that deflects a ray by an angle proportional to the radius of the beam in the lens. In the small angle approximation, such a lens will focus the beam (Fig. 1), with

$$\theta = x/f \qquad (1).$$

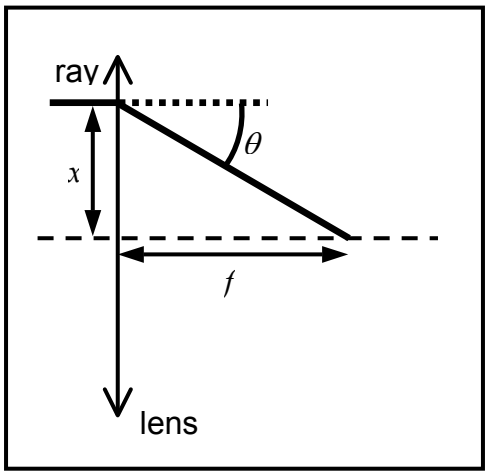

Figure 1. A schematic diagram of the focusing of a ray at radius x in a parallel beam by a lens to form a crossover a distance f (the focal length) from the lens.

A more exact form of Eq. (1) for the geometry in Fig. 1 is

$$\theta = \arctan(x/f) \simeq x/f - x^3/3f^3 \qquad (2),$$

where we have expanded to the third order term. In practice we are dealing with beams with a maximum convergence angle of 25 mrad, for which the third-order term in Eq. (2) will be equal to 5 μrad and we might reasonably expect to neglect it. In terms of the spherical aberration, however, this third order deviation in the angle corresponds to a coefficient of spherical aberration, C_S, equal to $f/3$, which is similar to the value that might be expected in a state-of-the-art objective lens. This analysis is valid for the infinite demagnification condition shown in Fig. 1; conditions of finite magnification require a slightly different third order term. This example shows how the C_S of a modern TEM objective lenses is commensurate with the small deviations from a Gaussian lens required by geometry, and is not the major optical defect it is often perceived to be.

Given that electron lenses are already of high optical quality, it is now clear that the corrections being made to the wavefront by a spherical aberration corrector are relatively small and need to be extremely precise. In order to function effectively, an aberration corrector also needs to compensate for the additional "parasitic" aberrations that arise from imprecise machining and assembly, and magnetic inhomogeneities. Practical aberration correction has only now become practical through the development of techniques for measuring and correcting the parasitic aberrations, which has itself required the development of computers fast enough to make such measurements in a practical length of time.

2. The performance of installed Nion correctors

There are now five Nion C_S correctors installed in VG dedicated STEM instruments operating in the field. Four of these machines are 100 kV instruments (one of which is operated at 120 kV) and one of which is a 300 kV instrument. The design and operation of the corrector has been described in previous papers [5,6]. As described above, the key to the performance of these correctors is the ability to measure and correct for the parasitic aberrations that arise once the C_S has been corrected. Our software measures the aberrations using Ronchigrams, as described in more detail in Ref. [6]. Tuning the microscope at the start of a session typically takes about 10 minutes and is an automated procedure.

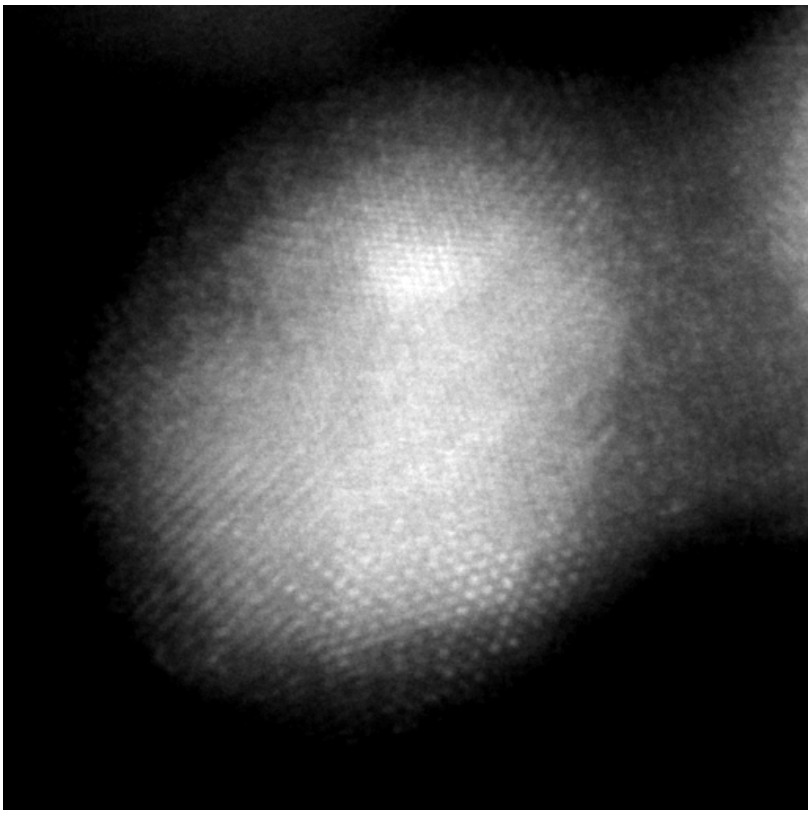

Figure 2. An HAADF image of a Au nanoparticle supported on amorphous carbon taken using the Nion corrected 100 kV VG STEM. The near horizontal fringes at the top of the particle have a spacing of 1.2 Å.

All of the 100 kV instruments have attained a resolution approaching or beyond 1 Å in HAADF images. Figure 2 illustrates how the resolving power of the microscope is improved. Prior to correction, the 2.3 Å {111} fringes in the <110> oriented domain at the bottom of the particle would just have been resolved. With correction, much smaller spacings can be observed in a domain at the top of the particle and {113} fringes with a spacing of 1.2 Å are observed with good contrast.

The corrector installed in the 300 kV HB603 STEM at Oak Ridge National Laboratory has allowed that machine to routinely image at a resolution of 0.8 Å [2]. Figure 3 shows a similar Au nanoparticle with the constrast enhanced to show the many single atoms near the edge of, and between, the nanoparticles. The ability to observe single atoms so close to a nanoparticle indicates that the probe is free of the long range tails that can occur if the imaging conditions are not optimized. An intensity profile of a pair of Au atoms show that they have a FWHM of 0.7 Å, indicating that the probe diameter is smaller than this (since the atom potential has a finite width). Note also that the profiles of the atoms both show a similar slight asymmetry in the probe, perhaps due to some small residual coma. The ability to image routinely at this resolution is a remarkable development, but measurement of the aberrations suggests that the aberration correction is sufficient for imaging at a resolution better than 0.6 Å. Such performance has yet to be achieved, and is probably being limited by instabilities of the microscope that have yet to be cured. The corrector cures spherical aberration but not instabilities, and it is remarkable that the resolutions described above have been

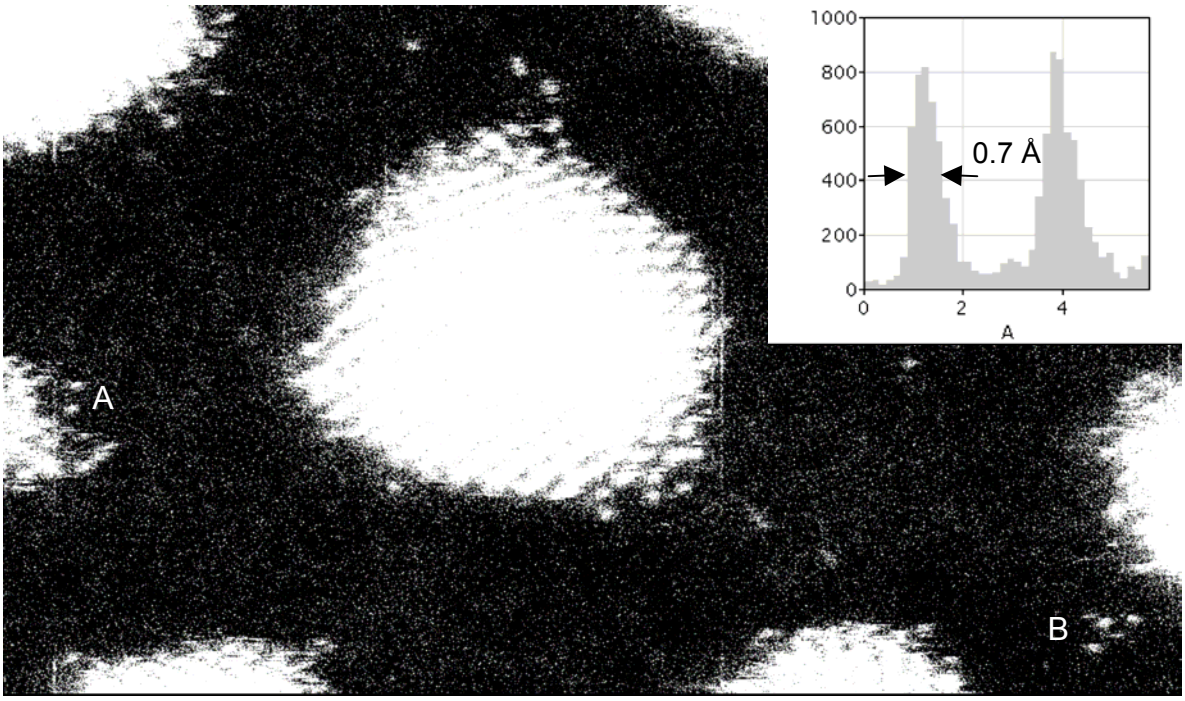

Fig. 3. An HAADF image of Au nanoparticles on an amorphous carbon film taken using the corrected 300 kV VG HB603 STEM at Oak Ridge. Inset: A profile through the pair of atoms indicated at A. The image of the atoms has a FWHM of 0.7 Å, and the atoms are separated by 2.8 Å. The trimer of atoms at B have similar dimensions.

achieved in columns that were not originally designed and specified for such performance.

3. EELS and spectrum imaging in a corrected STEM

In addition to improving the spatial resolution, a benefit of aberration correction that may turn out to be an even more epoch-defining advance is the increased beam current that becomes available. Correction of the C_S allows larger objective apertures to be used which, for no change in demagnification of the source, will lead to a significant increase in the probe current. In a 100 kV VG STEM with a cold field emission source, it is possible to focus more than 200 pA into a probe smaller than 1.4 Å. Previous to correction, 200 pA would only have been available in a probe of about 5 Å. It is likely that elemental mapping by electron energy-loss spectroscopy at 1-2 Å resolution will become possible in a minute or so for a 256 by 256 pixel elemental map.

Making use of the additional current for spectroscopy requires that the increased beam angles in the probe can be coupled into the spectrometer, which means that the spectrometer collection semiangle needs to be at least 20 mrad. Most VG instruments are not fitted with a post-specimen lens, and the additional optics can only easily be added after the lid of the machine, about 240 mm after the sample. Even assuming that the objective lens magnetic field provides a typical post-specimen compression of 3 times, a spectrometer at this position would require an entrance aperture of greater than 3 mm to collect the whole beam. At this size, the highest energy-resolution will not be available due to the aberrations of the spectrometer.

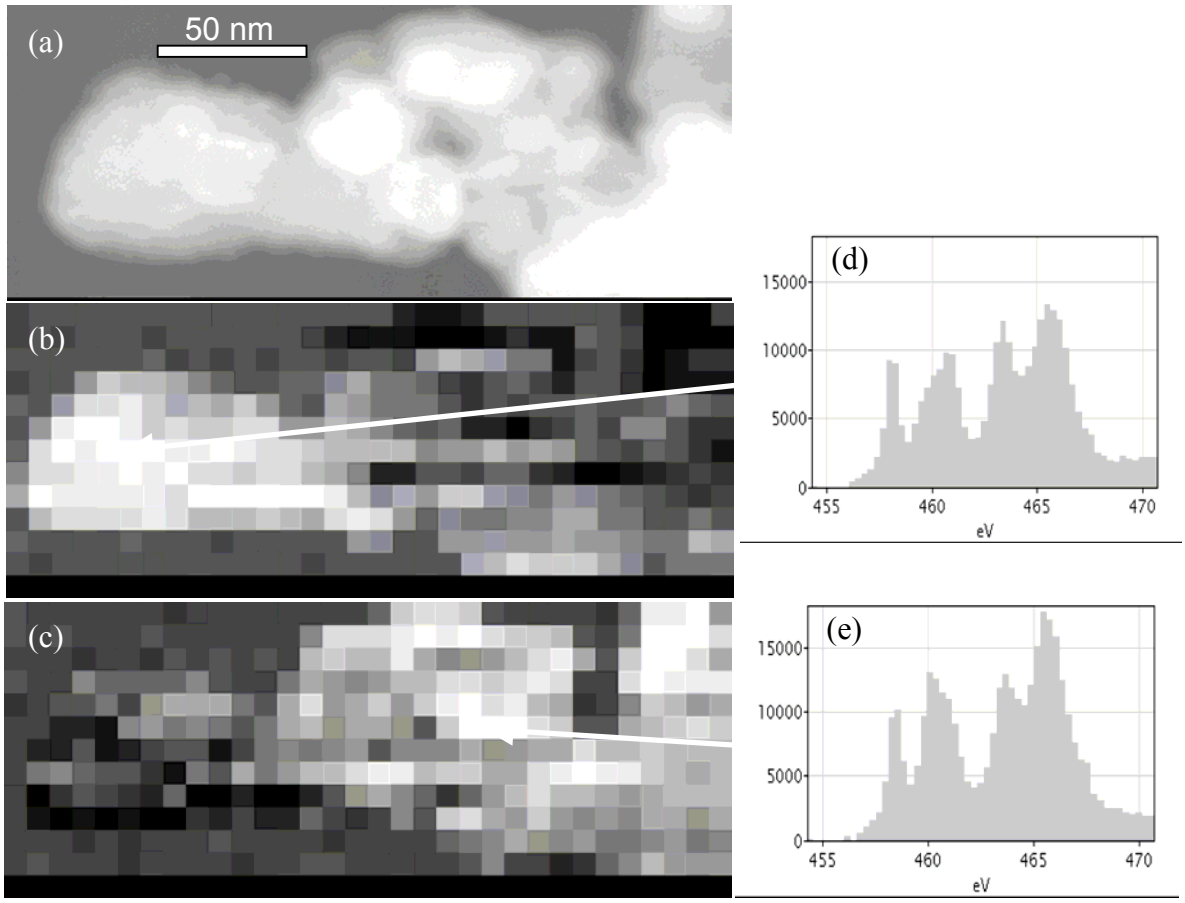

Figure 4. Data from a spectrum image (SI) of a TiO_2 catalyst support particle. Spectra were recorded on a 32 by 13 array of probe positions, with 0.5 s exposure per spectrum, 0.3 eV per channel dispersion. (a) The HAADF reference image of the particle. (b) and (c) The resulting images from a singular value decomposition (SVD) fit of (b) rutile and (c) anatase reference Ti $L_{2,3}$ spectra to the SI data. (d) and (e) Spectra from the pixels marked with the arrows showing the obvious changed to the fine structure.

At Nion we have developed a quadrupole coupling module that resembles an aberration corrector. It consists of 4 quadrupoles which can be used to provide increased compression of the beam prior to entry into the spectrometer while also ensuring that the virtual cross-over of the beam is at the height expected by the spectrometer to avoid it injecting second order aberrations. The similarity of the QCM to the aberration corrector goes further in that both a- and b-type (skew) octupoles can be placed at positions where the beam is either elliptical or round. This means that the third order aberrations of the spectrometer can be corrected, allowing larger physical entrance apertures to be used.

To demonstrate the ability to record spectra quickly in a spectrum image, Fig. 4 shows data recorded from a sample of TiO_2 that contains both the rutile and anatase phases. A 2 mm entrance aperture was used, which allowed a 20 mrad collection semiangle with the additional compression of the QCM. Individual spectra from the data cube can be easily identified as being either predominantly rutile or anatase simply by inspection of the fine structure. For a more automated map, a singular value

decomposition fit can be used using reference spectra to map the regions of anatase and rutile. Such data can be used to "colour in" an ADF image of the region of TiO_2 identifying the phases, but for monochrome reproduction we display two separate images here.

4. Discussion and future directions

The improvement in performance of the VG microscopes that have been fitted with Nion aberration correctors has been dramatic. Our experiences with these projects have, however, indicated a number of limiting factors that will prevent further significant improvement [5]. The most important of these are:

1. The construction of the VG objective lens prevents the placement of optics close enough to the objective lens to allow optical coupling of the corrector to the objective lens. Without this optical coupling, the correction of spherical aberration generates increased 5th order aberrations that limit the resolution to about 0.8 Å at 100 kV.
2. The condenser lenses of the VG are mechanically instable, particularly after excitation changes when they suffer from long term drift. In addition there are not enough electrical alignments provided to allow correct alignment.
3. The post-sample field of the objective lens is not strong enough to provide sufficient angular compression to allow large angle electrons to be collected into the EELS spectrometer without loss of energy resolution.

It is apparent that to make optimal use of the ability to correct the spherical aberration, a new column is required that addresses these issues. At Nion we are presently constructing such a column. A corrector has been designed that corrects 5th order aberrations in addition to the spherical aberration. The limiting aberration of this corrector is 8-fold astigmatism which is a 7th order aberration. In our design this is kept to about 1 cm, which would limit resolution at 100 kV to 0.4 Å. Of course, chromatic aberration will become increasingly significant at these resolutions. In our new column we have minimized C_C through the design of the objective lens and by minimizing the number of round lenses present in the probe forming optics. We anticipate that the C_C of the probe forming optics will be about 1.1 mm at 100 kV. Even so, achieving resolution better than 0.5 Å will probably require a 200 kV gun. The column is being designed for beams of up to 200 kV. Although the first column is to be mounted on an existing 100 kV VG gun, we are also engaged in developing our own 200 kV electron source.

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