A combined 200 kV HREM/STEM incorporating aberration correctors in both probe-forming and imaging lenses has been installed at Oxford University. This unique instrument is also equipped with an Ω-type in-column energy filter, annular dark field detectors both above and below the filter, and an EDX system. This paper presents an overview of the instrument together with data from initial performance tests that have demonstrated < 0.1 nm resolution in both TEM and STEM imaging modes.

Introduction

Transmission Electron Microscopy (TEM) is now clearly established as an invaluable tool in the determination of the structure and structural chemistry of a range of technologically important materials. In the last decade significant advances in instrumentation have been made with the development of Field Emission Gun (FEG) electron sources [1, 2] operating at intermediate (200-300 kV) voltages. These instruments provide imaging at close to 0.1 nm resolution, together with sub nanometer diameter bright probes which can be used to collect Electron Energy Loss Spectra (EELS) and to map characteristic X-Ray emission (EDX) peak intensities, providing chemical as well as structural information.

However, in the case of high resolution imaging, much of the extra information available can only be usefully extracted using indirect computational techniques such as exit wave reconstruction [3] or holography [4] due to the rapid oscillations in the wave aberration function at high spatial frequencies arising from the increased coherence of the source. For Scanning Transmission Electron Microscope (STEM) based imaging and analysis on the nm scale FEG sources are also indispensable providing increased probe current and reduced probe diameter. In parallel the development of energy-filtered imaging systems [5, 6] has enabled the intensity from electrons scattered inelastically by mechanisms other than phonon scattering to be eliminated from individual HREM images, improving quantitative interpretation and has enabled high resolution Electron Energy Loss Spectra (EELS) to be acquired.

A major practical challenge is now to further improve the limits of TEM and STEM resolution simultaneously by incorporating aberration correctors [7, 8] together with other analytical equipment, including energy filters in a single electron optical column. Recently this has been achieved and we present here the first of this new generation of aberration-corrected analytical FEGTEM/STEM instruments, recently installed at Oxford University’s Department of Materials. This unique instrument incorporates hexapole based aberration correctors in both the imaging and probe forming lenses [7, 8], together with an in column Ω type electron energy filter [5]. Pre and post filter High Angle Annular Dark Field (HAADF) detectors are also fitted, in addition to an EDX detector.

In this paper we first describe the instrument, and then present results from preliminary performance tests together with early results from various active areas of materials research at Oxford.

Design and Construction

The basic instrument (Figure 1) is based on the new JEOL JEM-2200FS column, incorporating a Schottky field-emission gun operating at up to 200 kV [2]. The aberration correctors are inserted into the column above and below the standard objective lens to enable aberration...
correction of both the illumination (CESCOR) and imaging (CETCOR) systems. Both correctors are based on a pair of strong hexapoles separated by a round doublet transfer lens enabling the correction of spherical and other aberrations to third order [7, 8]. In addition a series of additional weak multipoles are included within the correctors for adjustment of the beam axis and for the correction of residual parasitic aberrations. In order to ensure excellent mechanical and magnetic stability the electron optical column incorporates specially designed and improved magnetic coupling yokes and, as noted later, we have successfully maintained a stable corrected state over many hours. The electron energy filter is a corrected in-column Ω-type [5, 9] providing isochromatic imaging and diffraction together with high resolution EELS. Annular Dark Field detectors are fitted above and below the filter together with a conventional bright field STEM detector below the filter. An EDX detector (50 mm² thin window Si(Li)) is also attached to the column. The specimen stage is fully motorized on five axes and integrated piezo drives are provided for precise control of the specimen shifts.

The instrument is configured for full remote operation of the complete electron optical column including both correctors and all ancillary systems (apertures, detectors etc.) using five separate PCs communicating across a local area network and running a combination of Windows® and Unix® operating systems.

For remote viewing at TV rate a Hamamatsu (wide screen) camera system is used which includes automated black level, shutter and gain control, together with a Gatan Ultrascan 2024 × 2024 CCD camera coupled to an independent phosphor for high resolution image recording.

### Aberration Correction and Measurement

The Scherzer resolution [10] of the uncorrected objective lens (Cs = 0.5 mm) is 0.19 nm, measured from the calculated power spectrum of an image of a thin amorphous Ge film, calibrated from lattice fringes of Au particles with an information limit better than 0.14 nm. In order to obtain a corrected state, the tilt-induced values of 2-fold astigmatism and defocus are measured from a Zemlin tableau [11] of power spectra obtained from images recorded for a range of beam tilts. From these measured values the complete set of aberration coefficients to third order are calculated [12] and these values are used to apply appropriate voltages to the individual electron optical elements of the post imaging corrector.

In the corrected state, a typical set of calculated aberration coefficients are listed in Table 1. For images recorded in this condition the measured continuous signal transfer limits, as measured directly from the Young’s Fringe pattern shown in Figure 2(b), corresponding to 37% and 10% are 0.14(5) nm and 0.12(4) nm respectively. As the signal limit is now determined by higher order aberrations and effects of incoherent damping terms this data

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**Table 1**: Values of individual aberration coefficients to 5th order after correction of CETCOR.

<table>
<thead>
<tr>
<th>Coefficient</th>
<th>Aberration</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>2-fold astigmatism</td>
<td>239.3 pm</td>
</tr>
<tr>
<td>A2</td>
<td>3-fold astigmatism</td>
<td>33.5 nm</td>
</tr>
<tr>
<td>A3</td>
<td>4-fold astigmatism</td>
<td>639.5 nm</td>
</tr>
<tr>
<td>A4</td>
<td>5-fold astigmatism</td>
<td>18.2 μm</td>
</tr>
<tr>
<td>A5</td>
<td>6-fold astigmatism</td>
<td>1.626 mm</td>
</tr>
<tr>
<td>B2</td>
<td>1st order Axial coma</td>
<td>4.625 nm</td>
</tr>
<tr>
<td>B4</td>
<td>2nd order Axial coma</td>
<td>71.15 μm</td>
</tr>
<tr>
<td>S2</td>
<td>Star aberration</td>
<td>685.4 nm</td>
</tr>
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<td>D3</td>
<td>Defocus</td>
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</tr>
<tr>
<td>C3</td>
<td>Spherical aberration</td>
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</tr>
<tr>
<td>C5</td>
<td></td>
<td>-2.08 mm</td>
</tr>
</tbody>
</table>

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**Fig. 1**: The JEOL JEM-2200FS installed at Oxford showing major electron optical components and detectors. The local computer network controlling the microscope, correctors and ancillary devices is visible in the foreground.

**Fig. 2**: Power spectra calculated from high resolution images of an amorphous Ge foil in both (a) uncorrected (at the Scherzer defocus) and (b) corrected TEM states. In the corrected state the power spectrum illustrated was calculated from a pair of laterally displaced images of a thin amorphous Ge film and shows information transfer to 0.12(4) nm (marked with a circle), corresponding to 10% signal transfer.

**Fig. 3**: Ronchigrams recorded from the same specimen as Fig. 2 in both uncorrected and corrected STEM states.
also serves to confirm the excellent mechanical and electrical stability of the total system.

High resolution STEM imaging and analysis both require the smallest possible probe with the highest possible current. This has previously been successfully achieved in corrected, dedicated cold field emission gun STEM instruments using an alternative corrector design based on a combination of quadrupoles and octupoles [13]. However, this is now also realized for the first time in a combined TEM/STEM instrument using a hexapole based corrector.

The measurement and subsequent correction of aberrations in the illumination system requires a suitable specimen that contains high-resolution, high-contrast information across the whole field of view at an ADF STEM image magnification of ~×0.3 M. In order to calculate the aberrations present in the probe-forming lenses, successive pairs of images are recorded at over-focus and under-focus conditions for a tableau of probe positions. From such a data set the complete set of aberration coefficients to third order can be determined in an analogous fashion to that used for TEM correction and these values are used to apply appropriate voltages to the individual electron optical elements of the probe forming corrector.

Direct measurement of the aberrations present in the probe can also be made using a Ronchigram of an amorphous specimen recorded with a large condenser aperture (~100 mrad) to enable inspection of the size and symmetry of the features at high angles surrounding the central zone which reflect the symmetry of the residual, higher-order aberrations [14] (Figure 3). The present microscope has the unique advantage that the probe can also be imaged directly to measure the final corrected state. The potential for using aberration-corrected probes with convergence semi-angles up to 30 mrad is currently being investigated, since such probes have been calculated to contain up to an order of magnitude more current than probes of similar size in an uncorrected instrument employing the same source at the same accelerating voltage. We intend to exploit this in both EELS and EDX analysis at ultra-high spatial resolution.

**Initial Applications**

The microscope is fitted with a bright-field and an ADF STEM detector below the in-column energy filter to allow unfiltered or energy-filtered STEM images to be acquired. A second HAADF detector is positioned above the in-column filter so that a stationary probe position can be selected from within a scan to allow an EELS spectrum to be acquired from a selected point without any disturbance due to the insertion or removal of any aperture or detector. The angular acceptance ranges of both sets of detectors can be varied by changing the magnification of the intermediate and projector lenses. Figure 4(a) shows an ADF image of a <110> Si crystal, in which individual atomic columns, spaced 0.136 nm apart in projection, are clearly resolved. The power spectrum calculated from this image (Figure 4(b)) indicates the transfer of the {226} reflection corresponding to a spacing of 0.082 nm.

As already noted commercial electron energy filters are now available in several distinct configurations both with in-column [5, 9] geometries and as post-column imaging spectrometers [6]. In the instrument described here an isochromatic corrected, Ω-filter was chosen for both EELS, and for energy-filtered imaging and diffraction. Figure 5(a) shows a series of energy-filtered images of a Y-doped Si3N4/SiC ceramic, clearly revealing Y segregation at the grain boundaries which is important in determining the mechanical and other properties of the material. Figure 6 shows an example of a zero-loss CBED pattern recorded from a <111> Si single crystal, experimentally confirming a filter acceptance angle of > 80 mrad in the corrected state.

With the versatility offered by this unique
instrument at our disposal we are planning a wide range of experimental programs of which we present one example here. As part of an ongoing study of complex oxides we are now in the position to resolve clearly both the cation lattice and the anion sublattice in many of these compounds. Combining the benefits of TEM aberration correction with indirect specimen exit wave reconstruction from focal or tilt series [3] of aberration corrected images we can now achieve directly interpretable structural resolution below 0.1 nm. As an example, Figure 7 shows the restored exit wave of a complex niobium oxide block structure in which all atom positions, including those of oxygen, are visible in the restored phase and in which structural information has been recovered to 0.09 nm. It has also been possible to record ADF STEM images of the same crystal (Figure 7(b)) which do not indicate any notable segregation of cation species. We are currently extending these studies into further investigations of interstitial anion and cation distributions in these materials using both recovered specimen exit wave functions in combination with ADF STEM imaging.

Conclusions
A unique double aberration-corrected, energy-filtered analytical HREM/STEM has now been installed and tested at Oxford. Initial data from a range of materials has confirmed that it is capable of resolution below 0.1 nm in both HREM and STEM modes and it is now being used in a wide range of applications in materials science.

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References