The Aberration Corrected JEOL JEM-2200FS FEG-STEM/TEM Fitted with an \(\Omega\) Electron Energy-Filter: Performance Characterization and Selected Applications


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A newly developed JEOL JEM-2200FS scanning transmission electron microscope (STEM) / transmission electron microscope (TEM) integrated with an aberration-corrector and an in-column \(\Omega\)-filter has recently been installed at Lehigh University. For this instrument, all the microscope parameters, including room environment, have been optimized in order to achieve the best resolution in a STEM imaging mode while simultaneously allowing efficient analytical capabilities by electron-energy-loss spectrometry (EELS). The STEM and TEM performances of the Lehigh JEM-2200FS are presented in this report as well as some initial results of atomic-column STEM-EELS analysis obtained from a Ni-base intermetallic alloy.

Introduction

The identification and quantification of the nature of individual atomic configurations are among the ultimate goals for the characterization of nano-structured materials. It is now possible to perform such advanced characterization in a scanning transmission electron microscope (STEM) equipped with a recently developed aberration corrector using electron energy-loss spectrometry (EELS) and X-ray energy dispersive spectrometry (XEDS). The primary improvement due to the aberration corrector is a significant reduction of the effects of spherical aberration, which has been one of the major limiting factors of electron lenses since the original development of transmission electron microscopes (TEMs). By employing aberration correction, the contrast delocalization effect can be significantly minimized for atomic-resolution HAADF-STEM imaging in static-beam TEM applications [e.g., 1, 2]. In addition, the STEM aberration-corrector can refine the incident probe dimensions significantly. In aberration-corrected STEMs, therefore, the resolution in high-angle annular dark-field (HAADF) imaging can reach sub-

The Microscope Configuration

Figure 1 shows the JEM-2200FS 200 keV STEM/TEM installed at Lehigh University. This instrument is equipped with a Schottky...
field-emission gun (FEG) source and an in-column Ω-type electron energy-filter. In addition to the regular TEM functions such as bright/dark-field imaging, electron-diffraction analysis and atomic-resolution phase-contrast imaging, the Ω filter allows acquisition of zero-loss (fully elastic) images and elemental distributions in thin specimens using inelastically scattered electrons. In the previous JEOL Ω-filter microscopes, the illuminated area is significantly shrunk and distorted as the accelerating voltage is increased, which is required to acquire energy-filtering elemental maps. This change in the illuminated area is due to the strong pre-field above the specimen, which in combination with the change in the accelerating voltage made energy-filtering work more difficult. In the JEM-2200FS, however, the illuminated area in the energy-filtering TEM mode remains the same at any energy-loss region as a function of illumination compensation. In practical energy-filtering work, this illumination compensation feature is found to be a highly desirable and effective modification. Furthermore, this instrument is also equipped with a CEOS Cs-corrector in the illumination system (above the objective lens) for probe refinement.

In addition to these major distinct features, many upgrades have been made from previous JEOL Ω-filter instruments. The post-specimen column lens configuration have been modified so as to have as 4 and 2 post-specimen lenses at the pre- and post-Ω-filter positions, respectively. The additional post-specimen lens at the pre-Ω-filter position inhibits any image or defraction-pattern rotation as the magnification or camera length is changed. There is also a HAADF detector above the Ω filter, which makes STEM-EELS acquisition possible in combination with the additional post-specimen lens. This microscope is also equipped with another bright-field (BF) and ADF detectors positioned after all the lenses. In addition to the EELS functions facilitated by the Ω-filter, a ThermoElectron XEDS detector having a collection angle of 0.13 sr is also fitted to this microscope.

Microscope stabilities have also been improved. The specimen holder (which may be the most sensitive to influence by the external environment) is completely isolated by an o-ring sealed “clam shell” after loading into the microscope column. Due to the clam shell, stage drift is significantly reduced. The JEM-2200FS microscope is equipped with active suspensions at the four corners of the instrument base, which act to efficiently reduce high frequency vibrations.

The whole microscope is operated through different 5 computers (namely for (i) the major controls, (ii) STEM operation, (iii) image/EELS acquisition, (iv) XEDS acquisition and (v) aberration correction), which are linked via a local area network. Because all the controls, including apertures and energy-selection slit, can be accessed digitally, the instrument can also be operated remotely with the JEOL Sirius interface. For instance, the microscope has been operated remotely via a regular internet line from NASA Goddard, MD, USA, which is located ~280 km away from Lehigh University.

**The Room Construction**

To achieve 1 Å or sub-Å resolution in STEM imaging and analysis, the local environment surrounding the microscope needs to be carefully controlled. The key environmental factors that influence microscope performance can be summarized as: (i) stray electromagnetic fields, (ii) air flow, (iii) sudden air pressure changes, (iv) temperature drift and (v) residual noise (e.g. caused by fans in power supply units and computers). Some of these factors are strongly correlated with one another, and hence minimizing these environmental issues are essential requirement for advanced microscopy performance at atomic-column resolution levels. The microscope room for the JEM-2200FS at Lehigh has been modified from its previous configuration based largely on practical suggestions described by Muller and Grazul [6].

In order to reduce the electromagnetic fields, all metal parts such as ceiling grids, pipes, HEPA filters and duct work as well as metal raceways for wiring were removed. The major power supply units for the microscope were isolated in a separate air-conditioned room. All fluorescent lights were replaced with high intensity fluorescent bulbs and discrete sockets were installed for accessory power.

For the reduction of instabilities arising from air flow and air pressure changes, air conditioning filters and ducts were removed and a low-flow duct soc was installed on one of the walls at a distance of more than 3 m from the microscope column. In addition, an air-lock double door was installed and the microscope column has been surrounded with a curtain. The room temperature is controlled by 26 cooling panels mounted on the walls and ceiling as shown in Fig. 1. Since water circulated by a conventional chiller is used as coolant for the panels, no direct air flow is created for cooling unlike regular air conditioning. Using the cooling panels, the room temperature can be kept constant at 21°C with a temperature fluctuation of ~0.2°C per hour. For acoustic noise reduction, water chillers and vacuum pumps were positioned outside of the microscope room and acoustic baffles were installed between the cooling panels and ceiling/side walls as well as at each corner in the
room. In Fig. 1, the cooling panels and the acoustic baffles can be clearly seen. Furthermore, fan silencers were mounted on selected power supply units and computers to reduce noise.

**Characterization of Aberration-Corrected STEM Performance**

The CEOS aberration-corrector for the illumination system is positioned in the column between the condenser and objective lenses as shown in Fig. 1. The corrector consists of 2 hexapoles and 2 pairs of transfer lens couples, which can be tuned automatically using the Zelmin table method for probe shape analysis. This method involves measuring a series of probe shapes as a function of the tilt angle based on the deconvolution technique between focused and over/under-focused ADF images [7, 8]. After auto-tuning using the table method, a series of probe shapes at different tilt angles is displayed with the phase shift image being calculated from the measured aberration coefficients up to 5th order (Fig. 2). Typical values of the measured aberration coefficients after auto-tuning are also summarized in Table 1. Since the JEM-2200FS at Lehigh is equipped with an ultra high resolution pole-piece, the original values of the spherical ($C_s$) and chromatic ($C_z$) aberration coefficients, which are the smallest obtainable in current available 200 keV instruments, are shown in Table 1 for comparison. After aberration correction, the $C_s$ value is significantly reduced to 3.8 μm (1/120 of the original value!) with a 5th-order aberration coefficient ($C_5$) of only 3.2 mm. Note that the $C_s$ value in the corrected condition is slightly enlarged from the original value due to the presence of the aberration corrector. However, this enlargement in $C_s$ has only a marginal effect on the fine probe formation in a 200 keV FEG instrument.

After the auto-tuning process, lower order aberrations such as the twofold astigmatism ($A_2$) and 2nd-order axial coma ($B_2$) can be adjusted manually by observing the Ronchigram (i.e. the defocused shadow image). Figure 3 shows comparisons of the Ronchigrams obtained from Au nanoparticles on a thin Ge support film in the hexapole-off (a) and on (b) (i.e. aberration corrected) modes. It should be mentioned that the correction can be defeated by turning the hexapoles off even after correction tuning. In both Ronchigrams, the intensity distribution becomes uniform at the center. It is usually considered that the lens aberrations no longer influence the incident probe formation within the region of uniform intensity within the Ronchigram. In the uncorrected mode with $C_s=0.5$ mm, the uniform intensity area in Fig. 3(a) is only 11 mrad wide, and hence a smaller probe-forming aperture (which also limits the available probe current) must be used to avoid the influence of spherical aberration. Conversely, the uniform intensity region in the Ronchigram is expanded to over 24 mrad after corrector auto-tuning followed by the manual adjustments of $A_2$ and $B_2$, as shown in Fig. 3(b). The expansion of the uniform intensity area means that a larger probe-forming area can be selected for optimum imaging quality.

**Performance**

**CEOS aberration-correction system**

Table 1

<table>
<thead>
<tr>
<th>Condition</th>
<th>Spherical aberration, $C_s$ (mm)</th>
<th>Chromatic aberration, $C_z$ (mm)</th>
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<tr>
<td>(a) conventional</td>
<td>$C_s=0.5$</td>
<td>$C_z=1.1$</td>
</tr>
<tr>
<td>(b) aberration-corrected</td>
<td>$C_s=1.1$</td>
<td>$C_z=0.5$</td>
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</table>

<table>
<thead>
<tr>
<th>Aberration-type</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>Twofold astigmatism, $A_2$</td>
<td>$5.8$ mm</td>
</tr>
<tr>
<td>2nd-order axial coma, $B_2$</td>
<td>$40$ nm</td>
</tr>
<tr>
<td>Threefold astigmatism, $A_3$</td>
<td>$94$ nm</td>
</tr>
<tr>
<td>3rd-order spherical aberration, $C_3$</td>
<td>$3.8$ μm</td>
</tr>
<tr>
<td>3rd-order star aberration, $S_3$</td>
<td>$300$ nm</td>
</tr>
<tr>
<td>Fourfold astigmatism, $A_4$</td>
<td>$226$ nm</td>
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<td>4th-order spherical aberration, $C_4$</td>
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<tr>
<td>4th-order axial coma, $B_4$</td>
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</tr>
<tr>
<td>Fivefold astigmatism, $A_5$</td>
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<td>5th-order spherical aberration, $C_5$</td>
<td>$3$ μm</td>
</tr>
<tr>
<td>Sixfold astigmatism, $A_6$</td>
<td>$1$ mm</td>
</tr>
<tr>
<td>Chromatic aberration, $C_z$</td>
<td>$1.4$ mm</td>
</tr>
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</table>

Table 1 Summary of aberration coefficients of the JEM-2200FS for (a) conventional and (b) typical aberration-corrected conditions.

Fig. 3 Ronchigrams (defocused shadow images) recorded from a specimen of Au nano-particles on a Ge film at nearly focused conditions in the JEM-2200FS: (a) uncorrected mode with hexapole-off and (b) corrected mode with hexapole-on. The twofold astigmatism and 2nd-order axial coma were manually aligned.

Fig. 4 (a) The probe diameters containing 50% of the total intensity simulated based on the polychromatic point source for the conventional (open triangle) and aberration corrected (full aberrations: open circles, main aberrations only: closed circles) JEM-2200FS, plotted as a function of the convergence semi-angle $\alpha$. (b) The overall probe diameters containing 50% and 90% of the total intensity simulated by convolving the polychromatic intensity distribution with the Gaussian intensity distribution, plotted against the probe current. The polychromatic intensity distributions at $\alpha=12$ and 25 mrad were used for the conventional (open & closed triangles) and aberration-corrected (open & closed circles) conditions, respectively.

Fig. 8 (a) An HAADF-STEM image of an L1$_2$-type’ precipitate along <100> in the Ni-base superalloy X-750. This image is slightly distorted due to some instabilities during acquisition. (b) Intensity profiles extracted from atomic layers containing only face-centered atoms (A site) and both face-centered and corner atoms (A & B sites). (c) EELS spectra around the Ti L$_2,3$ edge measured from the A- and B-sites, respectively. (See the next page.)
ing aperture can be used without suffering any influence of lens aberration. Therefore, the use of aberration correction for probe refinement can provide not only improved resolution in HAADF imaging, but also actual benefits for analysis via X-rays or energy-loss electrons due to the significantly increased probe currents (5-10 \times 10^{-10} A).

Theoretical evaluation of probe dimensions in aberration-corrected STEM

Both imaging and analytical performance in STEM (as exemplified by resolution and analytical sensitivity) are directly influenced by the incident probe dimensions (i.e., sizes and intensity distributions). The finest probe with the highest current is always preferable to achieve the best imaging and analytical performance. In order to perform STEM imaging and analysis with the highest possible resolution and sensitivities, it is essential to estimate the optimized probe forming conditions and the probe dimensions for appropriate applications. Since the uniform intensity area in the Ronchigram is expanded by the STEM aberration-correction as shown in Fig. 3, the optimized conditions in the aberration-corrected mode are very different from those encountered in the conventional situation. In general, the complete probe simulation can be performed via three steps: (i) calculation of the intensity distribution from a monochromatic point source that contains only geometrical aberrations and defocus, (ii) calculation of the intensity distribution from a polychromatic point source including chromatic aberration by adding a series of monochromatic intensity distributions with certain weights based on the electron energy spread function of the gun and (iii) calculation of the intensity distribution from an overall extended source by convolving the polychromatic distribution with the contribution of the probe current. The details of the probe simulation are summarized by Colliex and Mory [9]. In this study, the intensity distributions from the monochromatic and polychromatic point sources were simulated by following an approach described by Haider et al. [10].

Figure 4(a) shows probe diameters of the polychromatic intensity distributions containing 50% of the total intensity (which is usually considered as a suitable size for HAADF image resolution) in the conventional and aberration-corrected conditions of the JEM-2200FS, plotted as a function of the convergence semi-angle (\( \alpha \)). For this simulation, the aberration coefficients summarized in Table 1 were used with the source energy spread of 1.0 eV. It should be noted that the probe diameters in the aberration-corrected conditions were simulated with only major aberration coefficients of \( C_5 \) and \( C_7 \) (closed circles) and with full aberration coefficients up to 5th-order except for \( A_1 \) and \( B_1 \) (open circles). Hence, it can be considered that the probe diameters simulated using only the major aberrations are ideal limits in this particular aberration-corrected condition. In the conventional condition, the probe diameter becomes a minimum (optimum) at \( \alpha = 11-12 \) mrad, which is consistent with the angular range of uniform intensity in the Ronchigram as shown in Fig. 3(a). In contrast, the probe diameter in the
aberration-corrected conditions becomes a minimum at $\alpha = 20-26$ mrad, which is higher than the optimum $\alpha$ in the conventional condition. However, as an estimation of the optimum aperture size, the aberration-corrected condition is still smaller than the angle measured from the uniform intensity area ($> 34$ mrad) in the Ronchigram shown in Fig. 3(b). The difference in the optimum angle between the simulated probe diameter and the experimentally obtained Ronchigram suggests that the uniform intensity area size may not simply be used as an estimation of the optimum aperture size, especially in the aberration-corrected condition. Further discussion concerning the optimum probe conditions will be published elsewhere [11].

The simulated probe diameters shown in Fig. 4(a) represent blurring of a point source due to the geometrical (monochromatic) and chromatic (polychromatic) aberrations. These probe diameters are useful to estimate the optimum conditions for obtaining ideal resolution of the HAADF-STEM imaging with limited probe currents. However, it is essential to take into account the contribution of the probe current in order to estimate spatial resolution for analysis via X-rays or electrons in the optimum conditions, as pointed out by Brown [12] and Watanabe et al. [5]. Because the contribution of the probe current can be described by assuming an initial Gaussian diameter at the electron gun, the overall probe diameter extracted from the extended source can be calculated by a convolution of the polychromatic intensity distribution with the Gaussian image of the electron gun [9]. For simplicity, the Gaussian diameter at a brightness value of $2 \times 10^7$ Am$^{-2}$sr$^{-1}$ was used for the Schottky FEG source [13]. The overall probe diameters simulated at the optimum $\alpha$ for the conventional and aberration-corrected conditions (12 and 25 mrad, respectively) are plotted against the probe current in Fig. 4(b). Probe diameters containing 90% of the total intensity (closed symbols) are also plotted in addition to the 50% diameters (open symbols) since the 90% diameter is a more relevant definition for chemical analysis [5, 14]. In the conventional condition, the probe current is usually limited to $\leq 10$ pA for atomic resolution HAADF-STEM imaging and the 50% and 90% probe diameters are 50 and 90 pA, respectively (plotted in red in Fig. 4(b)). The simulated intensity distribution of the incident probe and a HAADF-STEM image of Si $<$110$>$ recorded in the conventional condition are shown in Figs. 5(a) and 6(a), respectively. The image is very noisy due to the limited probe current available, and the projected Si-Si dumbbell spacing of 1.36 Å could not be resolved.

In the aberration corrected condition, the probe current can be increased to over 100 pA to obtain the same 50% probe diameter (1.6 Å) as in the conventional condition. This $\geq 10 \times$ higher current can improve the analytical sensitivity in XEDS and EELS experiments. On the other hand, the 50% diameter can be made smaller by using a probe of higher current, and the probe current range between 10-100 pA, all the microscope parameters such as the gun emission and gun/column lens setting were carefully chosen in order to achieve improved resolution in HAADF-STEM imaging and to gather sufficient signals for EELS analysis. Conversely, the energy filtering of the incident electrons needs to be kept as good as possible since the energy resolution is usually degraded as the gun emission increases to generate higher probe currents. Therefore, it has been found that the current at which the probe becomes converged, as promised by the above conditions, is around 50 pA. According to the results of the probe simulation shown in red symbols in Fig. 4(b), the diameters containing 50 and 90% of the total intensity are 1.2 and 3.5 Å, respectively.

**Figures 5(b) and 6(b) show the simulated probe shape and a recorded HAADF-STEM image of Si $<$110$>$ with 50 pA in the aberration-corrected condition. In the conventional condition, the corrected probe is about twice that in the conventional probe. This enhancement can provide a better signal-to-noise ratio in imaging. It is however important to note that the aberration-corrected probe has threefold tails instead of the ring-shape tail typical of a spherically aberrated probe (Fig. 5(a)). The threefold tails in the corrected probe are caused by the residual higher-order aberrations in the hexapole-based corrector. In comparison with the conventional image in Fig. 6(a), the ADF-STEM image of Si $<$110$>$ (Fig. 6(b)) is greatly improved with higher contrast and the 1.36 Å spacing is now successfully resolved. Figure 7 shows some EELS results obtained in the aberration-corrected STEM condition. Figure 7(a) presents the energy resolution in this particular condition determined by recording two zero-loss peaks separated with a 10-eV energy shift. Figure 7(b) shows an EELS spectrum in the vicinity of the Si K edge (1839 eV) for a 10-s acquisition. It is possible to measure such an EELS spectrum at higher core-loss regions with a reasonable acquisition time while still maintaining the atomic-scale resolution (1.36 Å) and a reasonable energy resolution ($\sim$1.1 eV). Therefore, atomic-column characterization of materials can in practice be performed by EELS analysis in the aberration-corrected JEM-2200F.

**A demonstration of atomic-column characterization**

*Figure 8(a) shows an atomic-resolution HAADF-STEM image recorded from a $<$100$>$ projection of an L1$_2$-type $\gamma$ precipitate (Ni$_2$Al) in a Ni-base superalloy (X-750). In this particular projection of the precipitate, the atomic separation is resolved by the EELS spectra obtained from the HAADF image. Furthermore, there are two distinctly different intensities displayed by the major atomic columns in the HAADF image. Intensity profiles extracted from an atomic layer containing only brighter atomic spots and from a layer with both brighter and darker columns in the HAADF image are shown in Fig. 8(b), respectively. The intensity at the darker columns are $\sim$1/3 of that found in the brighter columns, which is indicative of the difference in average atomic number between the projected atomic columns. In comparison with the expected atomic configurations in the L1$_2$ structure, the brighter columns correspond to face-centered Ni (A site), whereas the less intense columns correspond to Al (B site) atom positions in the L1$_2$ structure.*

According to XEDS analysis performed prior to the HAADF and EELS approaches, this $\gamma$ precipitate has a high Ti content (about twice that of Al in atomic fraction). *Figure 8(c) shows two EELS spectra in the vicinity of the Ti L$_{2,3}$ edge ($\sim$450 eV) recorded from the A and B sites, respectively. It is clear that the Ti L$_{2,3}$ edge intensity is more significant in the spectrum obtained from the B site. These results suggest that the Ti atoms preferentially occupy the B-site in the L1$_2$ structure by substituting for Al atoms, which agrees with previous studies using the X-ray based ALCHEMI method [e.g. 15]. Using the new generation aberration-corrected STEMs, therefore, such site occupancies of alloying elements or even impurity elements can be determined directly at the individual atomic-column level, not by averaging information from broad regions as is traditional.*

**TEM Performance with an Activated STEM Corrector**

In addition to the impressive STEM performance, the JEM-2200FS has all the TEM functionality as well, and it is designed so that both the TEM and aberration-corrected STEM modes can be seamlessly switched back and forth. TEM performance of this type has previously been reported for an instrument with a TEM aberration corrector [2, 8]. In the original TEM mode, the aberration corrector has been activated. In the JEM-2200FS at Lehigh, however, the TEM mode has been set up while keeping the aberration corrector activated. This special TEM mode is called the CsSTEM mode and is very important for doing atomic resolution STEM work where one wishes to avoid any electronic instabilities caused by turning the STEM corrector on and off. Several TEM-resolution performance characteristics in the CsSTEM mode are summarized in Fig. 9. The point resolution as determined from Au particles on a Ge thin-film using the optical diffraction method is 0.19 nm (Fig. 8(a)). The information limit measured from a polymer-crystalline sample was determined by an energy-loss method and was found to be 0.11 nm (Fig. 9(b)).

*Figures 9(c) and d show an atomic resolution phase contrast image from Si $<$110$>$ and its Fourier-transformed diffraction pattern, respectively. Although the 1.36-A atomic spacing is not resolved in the phase contrast image, Bragg spots corresponding to $[224]$, [115] and [333] with those in the original TEM mode, it can be concluded that the TEM performance of the JEM-2200FS is not degraded at all by the activation of the STEM corrector. Additionally, a static convergent probe can also be formed with STEM-corrector activation, which is called the CsCBD mode. In contrast to the CsSTEM mode, aberration correction is not actually corrected by the STEM corrector. Therefore, the aberration-corrected fine probes*
in this instrument are available even in the static probe mode in addition to the scanning probe mode.

In the static probe mode, materials can also be characterized by energy-filtering via the Ω filter. Figure 10 shows a series of energy-filtered images taken from a cross-section Si/Si-Ge multilayer thin film, which is reported to be one of the most useful specimens for evaluating energy-filtered images [16]. Unfiltered (conventional) and zero-loss filtered bright-field (BF) images are shown in (a) and (b), respectively, and the relative thickness map determined from both the BF images is presented in (c). The elemental maps of Si L (d), Ge L (e) and Si K (f) ionization edges were obtained by the three-window method. These filtered images were recorded in the CsTEM mode. The elemental distributions using higher energy-loss edges such as the Ge L (1217 eV) and Si K (1839 eV) are very clear. This is a clear demonstration that the illumination compensation works very well at higher energy-loss regions.

Summary

The newly developed JEM-2200FS STEM/TEM has been successfully installed at Lehigh University. This instrument has been optimized for EELS analysis in the atomic-resolution HAADF-STEM imaging mode by the integration of the CEOS aberration-corrector for STEM and an in-column Ω-filter, which provides significant improvements in the imaging and analytical performance. Characterization on the atomic column-by-atomic column scale can be achieved using the JEM-2200FS with the STEM aberration corrector. In addition to the STEM mode, static-beam TEM-based operations can also be performed without any degradation in atomic resolution phase-contrast imaging or energy filtering in the STEM-corrector activated mode.

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References