Performance of a cold-field emission gun double aberration corrected TEM/STEM at 80kV

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Abstract: In this report we demonstrate the successful 80kV alignment of a double aberration corrected TEM/STEM (JEM-Z3100F-R005) equipped with a cold field emission gun designed to operate at 300kV. In TEM mode we demonstrate information transfer out to 0.078nm in the corresponding power spectrum. For high angle annular dark-field STEM, direct resolution of the 0.082nm Ge (444) dumbbells was achieved. The energy spread of the primary electron beam was studied using energy-loss spectroscopy. Reduction of the extraction anode voltage (A1) to 1.2kV results in an energy resolution in the primary electron beam, taken as the full width half maximum (FWHM) of the zero loss peak, of 0.28eV with a corresponding beam current of 12pA whereas operation of A1 at 1.5kV results in a FWHM at the zero-loss peak of ~0.4eV for a beam current of 1.1nA.

1. Introduction
Lower operating voltages in the transmission and scanning transmission electron microscope (TEM/STEM) are particularly desirable to reduce the effects of knock-on damage during the study of beam sensitive materials such as those based on carbon and in particular the study of graphene [1-4]. Lower operating voltages also offer the potential for improved sensitivity energy-loss spectroscopy and X-ray spectroscopy due to the increased ionization cross-section. However, at lower accelerating voltages the resolution is reduced and chromatic aberrations are increased due to the longer wavelength of the incident primary electrons. Recent attempts to overcome these limitations have applied a combination of a cold field-emission gun (c-FEG) source and aberration correction of the probe and image forming lenses [5,6].

In this report we present the first results obtained at 80kV from a JEOL Z3100F-R005 (R005) TEM/STEM nominally designed to operate at 300kV. This instrument is equipped with a cold field emission gun consisting of a W <111> nano-tip emitter [7] and JEOL probe (STEM) and imaging (TEM) lens correctors. An asymmetric corrector configuration is utilized consisting of two dodecapole assemblies of different thicknesses coupled via transfer lenses of different focal lengths. Such an asymmetric corrector design has the advantage of suppressing chromatic aberrations otherwise introduced by symmetrical corrector designs and also reduces the effects of higher order parasitic aberrations [8].
2. Experimental methods

The c-FEG was operated at 80kV by utilisation of appropriate gun shorting electrodes to ensure the optimum position of the first cross over of the electron beam with respect to the subsequent condenser optics. Residual aberrations in the imaging and probe forming lens were minimised using the diffractogram tableau (Zemlin tilt-tableau) [9] and segmental Ronchigram auto correlation function matrix method respectively [10]. STEM high angle annular dark-field (HAADF) images of Si (110) and Ge (112) were obtained using a convergence semi-angle of ~28mrad and an inner and outer annular dark field detector angle of 86mrad and 229mrad respectively (nominal 15cm camera length). The energy resolution of the 80kV primary electrons was assessed as the FWHM of the zero energy-loss peak in vacuum as a function of the accelerating anode (A1) between 1.15 and 1.70kV (A2 held constant at 4.6kV) using image coupled mode (TEM spot size 1,2). The defocus of the illumination was adjusted using the C3 lens (brightness) to give a maximum ZLP intensity of ~60000 counts in each case. Spectra were obtained using a post column GATAN Tridium GIF™ with a 1mm GIF entrance aperture, 0.02eV/pixel dispersion and an exposure of 0.1 and 1.0 seconds respectively.

3. Results and Discussion

Typical residual aberrations for both the probe (STEM) and imaging (TEM) correctors at 80kV, after routine alignment using the automated corrector routine, are given in Table 1. Final adjustment of the 2-fold astigmatism $A_2$ and (for STEM) the axial coma $P_3$ is normally performed manually using the appropriate corrector deflector coils and the condenser lens 2nd beam deflector (bright tilt) respectively. The residual aberrations at 80kV are of a similar magnitude to those under optimised conditions at 300kV (also listed in Table 1 for comparison) with the exception of 80kV probe correction which shows a general increase of the fifth order Cs and six-fold astigmatism values. This is perhaps to be expected since the influence of fifth-order 6-fold aberrations ($A_6$) can become significant at lower operating voltages and the asymmetrical dodecapole-type aberration correctors of the R005 cannot compensate for 6-fold astigmatism [5].

| Aberration Coefficient | 80kV TEM | STEM | 300kV TEM | STEM |
|------------------------|----------|------|-----------|------|
| Defocus: $O_2$         | -223.18nm| 1.60nm| -226nm    | 3.81nm|
| 2-fold astigmatism: $A_2$| 6.03nm   | 21.40nm| 6.3nm     | 0.68nm|
| Axial coma: $P_3$      | 190.01nm | 213.61nm| 87.4nm    | 27.97nm|
| 3-Fold astigmatism: $A_3$| 33.50nm | 105.27nm| 60.3nm    | 7.70nm|
| Spherical aberration: $O_4$| -0.67µm | 3.28µm| 0.516µm   | 0.219µm|
| Star aberration: $Q_4$ | 1.22µm   | 2.03µm| 0.298µm   | 0.36µm|
| 4-fold astigmatism: $A_4$| 1.45µm   | 1.48µm| 3.05µm    | 0.80µm|
| Fourth-order axial coma: $P_5$| 111.31µm| 123.68µm| 6.01µm    | 32.24µm|
| Three-lobe aberration: $R_5$| 4.25µm   | 99.19µm| 30.24µm   | 6.07µm|
| 5-fold astigmatism: $A_5$| 37.15µm  | 95.03µm| 49.84µm   | 9.81µm|
| Fifth-order spherical aberration: $O_6$| -0.37mm | -2.05mm| -0.614mm  | -0.54mm|
| 6-fold astigmatism: $A_6$| 0.22mm   | 3.20mm| 0.78mm    | 0.41mm|

Table 1: Typical coefficients of the residual aberrations measured at 80kV and 300kV for TEM and STEM after aberration correction using the automated correction routine (note: the large TEM defocus value is a necessity of the correction routine).

Fig. 1(a) shows a high resolution TEM image from a Si [110] single crystal while the corresponding fast Fourier transform (FFT) indicates information transfer out to the Si (444) reflection, corresponding to 0.078nm. The Zemlin tilt-tableau generated to calculate the aberrations within the image forming optics is shown in Fig.1(b) with a maximum tilt angle of 45.25mrad. Fig. 1(c) shows a typical Ronchigram after probe correction from an amorphous film recorded by a 2k x 2k CCD camera (Gatan Ultra-Scan) mounted within the camera chamber. The Ronchigram shows a flat contrast region in the central circle with a half-angle of 38.8mrad compared to the typical flat region in the
Ronchigram at 300kV of 45-50mrad. STEM resolution is demonstrated in Fig. 2, (a) showing a lattice resolved HAADF image from Si [110], (b) the corresponding FFT showing transfer out to the (2-2 4) reflection and (c) the resolved 0.136nm dumbbell spacing. By careful adjustment of the corrector alignment to suppress the fifth order spherical aberrations to a minimum it has been possible to just resolve the (444) dumbbell spacing in Ge [112] with a contrast minima \( \sim 27\pm2\% \) of the maximum contrast intensity (after background subtraction) as illustrated in Fig. 3.

A plot of the energy spread of the electron source at 80kV, measured as the FWHM of the zero energy-loss peak, as a function of the extraction anode voltage (A1) is presented in Fig. 4(a). A minimum energy spread of 0.28eV was achieved with an A1 setting of 1.20kV (shown in Fig 4(b)). Further reduction of the extraction anode resulted in no further improvement in energy resolution until below an A1 of 0.9kV a rapid reduction in emission occurs with insufficient beam current to record a meaningful energy-loss spectrum. Under the conditions used to generate the zero-loss peak shown in Fig. 4b the emission current was significantly lower than 1\( \mu \)A to minimize the Boersch effect. The peak shape has a distinctive asymmetry with a broader tail on the higher energy-loss side. This effect was also observed in the minimum energy-spread ZLP at an operating voltage of 300kV (A1 at 0.9eV, emission current <1\( \mu \)A, FWHM 0.32eV). Increasing A1 to 1.70kV results at 80kV a corresponding
increase in the energy spread to 0.6eV FWHM as shown in Fig. 4c. The equivalent energy spread at 300kV for an A1 of 1.7kV is 0.95eV FWHM.

Fig 4: (a) Plot of primary electron beam energy spread as a function of extraction voltage (A1), (b) zero-loss peak (ZLP) for A1=1.20kV with a FWHM of 0.28eV and (c) corresponding ZLP for an A1 of 1.7kV with a FWHM of 0.6eV. The corresponding emission current for an A1 of 1.20kV and 1.70kV was <1 µA and 4 µA with beam currents of 12pA and 10.7nA respectively.

4. Summary
Sub-Ångström resolution has been successfully demonstrated at 80kV in a double aberration corrected c-FEG TEM/STEM designed to operate at 300kV. For TEM imaging, information transfer out to 0.078nm has been observed in the FFT of Si [110] while direct evidence of HAADF STEM resolution of the Ge (444) dumbbells of 0.82nm has been recorded. A best energy spread of the primary electron beam of 0.28eV was given for an extraction voltage of 1.20kV with an emission current of <1 µA.

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