Overcoming the chromatic aberration resolution limit by monochromation

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The quest for improved spatial resolution in aberration-corrected scanning transmission electron microscopy (STEM) resembles a set of Matryoshka dolls; as the resolution limit due to a particular aberration is removed, it reveals the next limiting aberration, typically of higher order. The first principal aberration to be removed was spherical aberration, in the 1990s. The limits then became different for quadrupole-octupole (QO) and sextupole-round lens (SR) correctors. For QO correctors, the next limit was 4-fold astigmatism of fifth order $C_{5,4}$, overcome by third-generation correctors in the 2000s. For SR correctors, the next limit was six-fold astigmatism $C_{5,6}$, minimized by shortening the sextupoles, and eliminated by three-sextupole correctors. The next limit is chromatic aberration C_c for both types of correctors. When it is overcome, 7th order and 6th order aberrations will become the main limit to overcome for QO correctors SR correctors respectively, provided that parasitic aberrations are sufficiently minimized.

The effect of chromatic aberration increases more rapidly than the effect of geometric aberrations as the primary energy E_o is lowered, which means that it is especially serious for STEM (or TEM) imaging at E_o<<100 keV. The C_c limit can be overcome with a chromatic aberration corrector, or by reducing the energy spread δE with a monochromator.

Fig. 1 shows a pair of experimental out-of-focus Ronchigrams recorded at 30 keV, without and with monochromation. Changing the beam energy slightly and measuring the defocus change showed that the STEM's total C_c was 0.97 mm. The full width at half maximum (FWHM) δE of the energy distribution in the non-monochromated case was 0.32 eV, and the corresponding defocus change $\Delta Z = C_c\, \delta E / E_o$ = 10 nm. The resultant Ronchigram is an incoherent superposition of shadow images of the sample taken at different magnifications, with clearly visible radial blurring. If this region near the edge is not cut out by the aperture, it contributes to the probe tail rather than the probe maximum and thus produces a loss of resolution and contrast. Cutting it out leads to poorer diffraction limit and hence decreased spatial resolution. A better solution is to decrease δE to 0.11 eV by monochromation, which narrows the defocus spread to ±1.5 nm and eliminates the radial blurring (Fig 1b).

Fig. 2 shows a single scan STEM HAADF image of graphene as acquired at 30 kV with δE = 0.1 eV with the third ring of graphene reflections and hence spatial resolution of 1.07 Å, and 50% modulation between the atoms and structure "holes". The probe current was about 10 pA. Without the monochromation, the probe current would have been ~30 pA and the resolution ~1.7 Å. The same improvement carried to 60 and 100 keV STEM imaging is expected to give resolution ~0.8 Å at 60 keV, and ~0.6 Å at 100 keV.

Monochromating has the additional advantage that the EELS energy resolution is greatly enhanced, to 5 meV in our monochromated EELS system at 30 keV. This opens up a large range of new experimental possibilities for electron spectroscopy.

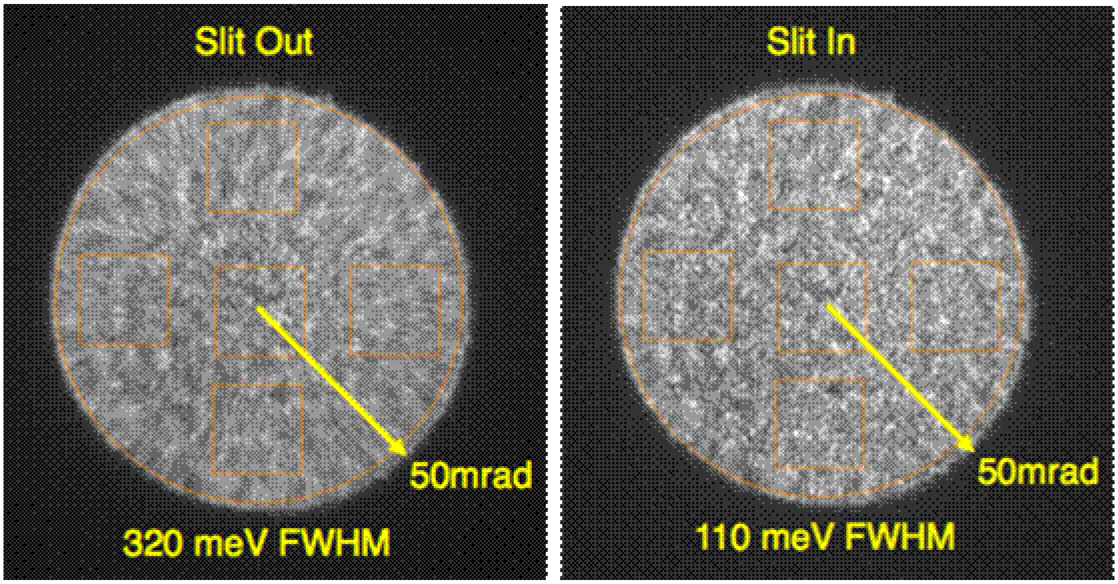

Fig 1 (a) (left) Ronchigram of amorphous carbon E_0=30keV, C_c=0.97mm, δE=0.32eV. (**b**) (right) Same as in (**a**) except δE=0.11eV

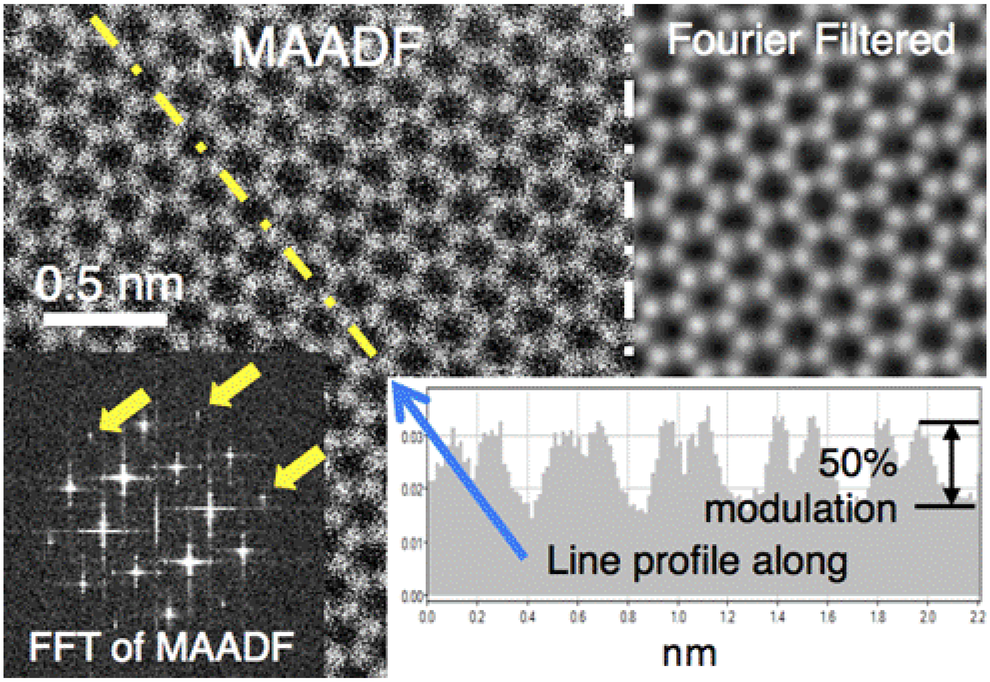

Fig 2. Medium-angle annular dark field (MAADF) STEM image of graphene obtained at 30keV, with FFT and a line profile taken along the indicated direction. E_o = 30keV, C_c = 0.97mm, δE = 0.11eV. Arrows in the FFT point to strongly transferred [20-20] 1.07 Å graphene spacings.