Aberration-corrected precession electron diffraction

C.S. Own,* N. Dellby,* O.L. Krivanek,* L.D. Marks,** and M. Murfitt

* Nion Co., 1102 8th St, Kirkland, WA 98033
** Northwestern University, 2220 Campus Dr., Cook 2036, Evanston, IL 60208

Precession electron diffraction (PED) is a promising technique for collecting high quality diffraction patterns for rapid nanoscale structural characterization [1]. It is able to reduce dynamical scattering effects, improving the interpretability of diffraction intensities over those obtained by conventional electron diffraction techniques. When used on a microscope that can produce a fine probe, the method simplifies symmetry identification and enables more straightforward phase recovery (using statistical inversion techniques) for small phases on the order of tens of nanometers. Several studies have reported remarkable improvements to dataset quality: $R$-factors of just above 10% have been reported for fairly thick metal oxide phases solved using PED data, as compared to 20%-40% typical for conventional selected area patterns (for reference, typical synchrotron X-ray $R$-factors are under 10%) [2-6].

The geometry of PED is shown in Fig. 1. The electron beam is tilted off-zone by angle $\phi$, and precessed azimuthally about the zone axis by $\theta = 2\pi$, forming a hollow cone of illumination. The scattered beams are scanned in a complementary fashion to restore a zone axis pattern, giving spots that are an incoherent summation of the constituent tilt components.

Whereas conventional diffraction is unaffected by the aberrations in the objective lens, precession diffraction poses a different set of instrument constraints where optical aberrations are quite relevant. These constraints are clarified by examining some of the features desired for PED:

1. Nanoprobe in the sample plane (~1 nm diameter)
2. Well-defined spots in the PED pattern
3. Minimal distortions in the PED pattern
4. Large cone semiangle (>60 mrad)
5. Continuously variable cone semiangles without need for major realignment

Features 1-3 are clearly needed for nanoscale study and easy dataset quantitation. Because of the large tilt angle and dynamic illumination during precession, the misprojection of the beam onto the sample plane by the aberrated objective lens pre-field causes the probe to wander and change shape, giving a delocalized precessed probe. Careful balancing of aberrations is needed to produce a nanoprobe in the microscope; indeed, keeping the probe stationary and of constant shape in PED is aberration correction by another name.

For large $\phi$, the smallest probe in an uncorrected instrument is obtained by adjusting defocus to compensate spherical aberration ($C_s$) to give an annulus of flat phase in the aberration surface $\chi$, similar to Scherzer defocus but with large defocus values of several $\mu$m. A side effect, however, is introduction of astigmatism ($C_{12}$) with direction and amplitude dependent on the defocus and $\phi$. This limits a probe in the same machine to one on the order of 10 nm at $\phi = 30$ mrad for 200
kV electrons. To achieve the desired 1 nm probe, $\phi$ must be less than 15 mrad. At 60 mrad, one is exploring regimes of high curvature in $\chi$ and the probe consequently becomes several tens of nm in size. Apart from $C_s$, the threefold astigmatism term dominates in conventional instruments since most machines do not contain a sextupole stigmator (Fig. 2).

In past implementations, we compensated the probe wandering by applying distorted scan waveforms so that the objective lens always sees a pre-aberrated pencil of illumination throughout the precession [7]. This was effective at providing a fairly localized probe ($\sim$15 nm), however, at high angles the aberration function $\chi$ varies tremendously, requiring re-tuning when $\phi$ increases beyond about 20 mrad. Although it is known that larger cone angles are better at reducing dynamical effects [8], a recent study shows that smaller angle PED is more sensitive to charge density effects [6]. Therefore, a continuous range of available cone angles is desired. Any of these should be available without need for re-tuning, however, this is not possible with conventional instruments.

Precession diffraction lends itself naturally to aberration corrected systems, which allow precise tailoring of the phase surface for forming highly convergent fine probes (a $C_3$ corrector can theoretically correct to about 60 mrad half-angle). A corrected system can thus provide a wide range of precession angles without the need for numerous pre-compensated waveforms and dynamic astigmatism correction.

The scan system of the Nion UltraSTEM™ is capable of providing precession in an aberration-corrected environment. PED on this system benefits from a symmetrical objective polepiece and scan/descan coils, and a $\pm 30^\circ$ compucentric double-tilt stage that provides eucentric tilts and reproducible mechanical adjustment on the order of nanometers (described elsewhere in the proceedings). Further details and results from this system will be reported at the meeting.


Fig. 1. Precession diffraction geometry.

Fig. 2. Misprojection of the precessed beam by a three-fold aberration function (left) generates a star pattern in the sample plane. Image taken on JEM-2000FX. $\phi = 40$ mrad, $d_{lobe} \sim 25$ nm.