

Utility of Precession Electron Diffraction Patterns with Varying Degrees of Non-parallel Illumination from the Same Nominal Sample Area

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Diffraction of a primary electron beam that is precessing, i.e. Precession Electron Diffraction (PED), in a Transmission Electron Microscope (TEM) [1-8], has over the last twenty-odd years become “*an invaluable tool, not only for the determination of unknown crystal structures but also in aiding the analysis of local microstructure*” [8]. Seven years after its first demonstration [9], the “scanning version of PED” [4-6] rivals in popularity electron backscattering in a scanning electron microscope.

The precession axis of the primary electron beam coincides *ideally* with the optical axis of the TEM and the (sharp) apex of the resulting hollow electron cone intersects the sample at a singular point. This ideal is, however, for a variety of reasons not readily achievable in practice. When the spherical aberration of the objective lens of the microscope is not corrected for and the alignment is not perfect, more or less blunt apexes of electron cones (that may not be completely hollow and round) result in PED. These cones intersect the sample over some area (rather than in a single point) and define the spatial resolution of the PED technique. The cones have also been observed to “wander” (or “wobble and dance” [7]) over some sample area [3,4]. Correction of spherical aberration of the (probe forming) objective lens pre-field may reduce this “wandering” to approximately 1 nm [7].

Utilizing the recently derived relation $t_{loc} \approx \frac{\lambda}{2} \langle (g_0(\alpha + \beta))^2 \rangle - \langle \alpha + \beta \rangle \langle g_0(\alpha + \beta) \rangle$, where t_{loc} is the local crystal sample thickness, λ is the electron wavelength, g_0 is the magnitude of the reciprocal lattice vector that determines the radius of the zero-order Laue circle, α is the precession half angle (5 – 60 mrad), and β is the convergence angle (0.5 – 5 mrad) [10], one can in principle assess the size of these sample areas. This is because the radii of the observed zero-order Laue circles in PED patterns must for a plane-parallel sample plate fall on an “equal thickness” curve in Figure 1. Observed deviations from this functional relationship are indications that the sample is not a plane parallel plate and that the apex of the electron cone is blunt. Utilizing wedge shaped samples (that often arise naturally by the crushing of inorganic crystals), one can then assess the illuminated sample area (as a function of the precession angle) after one has determined the wedge angle by some appropriate technique such as the spacing of two-beam extinction fringes. (The linear increase with convergence angle and the quadratic increase with precession angle [1,7,8] of the size of a blunt electron cone apex intersection with the sample may need to be considered in this context.)

In ref. [10] we proposed to utilize this relation (and its generalizations to higher order Laue zones) for the determination of those reflections that can be utilized straightforwardly for structural fingerprinting from PED patterns in a TEM [2]. That idea is based on Boris Vainshtein’s critical thickness (t_{cri}) criterion for quasi-kinematic diffraction: $\frac{\Omega}{\lambda} \cdot t_{cri} \leq |F_{hkl}|$, where Ω is the volume of the crystal’s unit cell and $|F_{hkl}|$ is the square root of a reflection’s intensity (i.e. a kinematical structure factor amplitude) [11]. Large precession angles lead generally to instantaneous “few-beam” conditions [2] that are better approximations to exact two-beam conditions (for which Vainshtein’s inequality has been derived originally). For candidate crystal

structures, Ω and $|F_{hkl}|$ will be known so that one needs to make sure that conditions of $t_{loc} \leq t_{cri}$ are fulfilled for those reflections that one utilizes for structural fingerprinting from PED patterns in a TEM (with fixed λ).

In summary, observed Laue circle radii in PED patterns as function of the degree of non-parallel illumination can be utilized on the basis of our ideas for both determination of the local sample thicknesses and assessments of the illuminated sample areas. Quasi-kinematic structural fingerprinting of nanocrystals from selected PED spot intensities is also possible on the basis of our ideas. As the precession angle magnitude has so far been rarely exploited as an experimental degree of freedom and because locally determined sample thicknesses are quite valuable, it is safe to predict that these kinds of ideas will prove their worth in the future in a range of TEM investigations of crystalline materials (including 6D electron tomography [12], which covers direct and reciprocal space simultaneously) [13].

- [1] R. Vincent and P. Midgley, *Ultramicroscopy* **53** (1994) 271.
 [2] P. Moeck and S. Rouvimov, *Zeitschrift für Kristallographie* **225** (2010) 110, open access: <http://www.degruyter.com/view/j/zkri.2010.225.issue-2-3/zkri.2010.1162/zkri.2010.1162.xml>
 [3] Y. Liao and L. D. Marks, *Ultramicroscopy* **117** (2012) 1.
 [4] P. Moeck *et al*, Proc. 11th IEEE International Conference on Nanotechnology, August 15-18, 2011, Portland, Oregon, USA, pp. 754-759, DOI: 10.1109/NANO.2011.6144300
 [5] P. Moeck *et al*, *Cryst. Res. Technol.* **46** (2011) 589.
 [6] A. S. Eggeman and P. A. Midgley, *Adv. Imaging Electron Phys.* **170** (2012) 1.
 [7] A. S. Eggeman *et al*, *Zeitschrift für Kristallographie - Crystalline Materials* **228** (2012) 43.
 [8] P. Midgley and A. Eggeman, *IUCrJ* **2** (2015) 126; open access: <http://journals.iucr.org/m/issues/2015/01/00/ro5004/ro5004.pdf>
 [9] E. F. Rauch *et al*, *Microsc. Anal.* **22** (2008) S5.
 [10] P. Moeck *et al*, *Microsc. Microanal.* **18** (Suppl. 2), 2012, p. 562.
 [11] B. K. Vainshtein, *Fundamentals of Crystallography, Symmetry and Methods for Structural Crystallography* (Springer, 2nd edition, 1994) p. 345; see also B. K. Vainshtein, *Acta Cryst. B* **47** (1991) 145.
 [12] A. Eggeman *et al*, *Acta Cryst. A* **70** (2014) C368.
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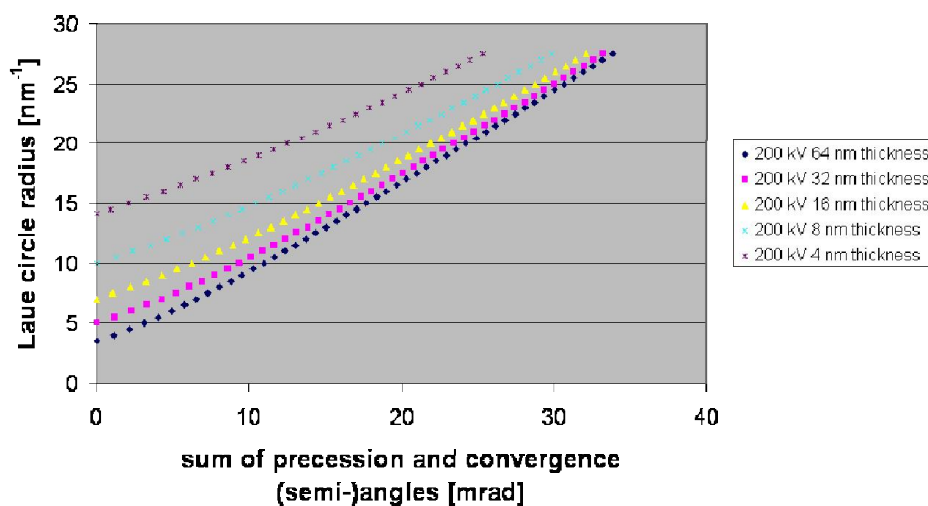


Figure 1. Predicted zero-order Laue circle radii in PED patterns versus degree of non-parallel illumination for plane parallel crystal plate samples with thicknesses between 4 and 64 nm.