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System Design and Verification of the Precession Electron Diffraction Technique

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ABSTRACT

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Bulk structural crystallography is generally a two-part process wherein a rough starting structure model is first derived, then later refined to give an accurate model of the structure. The critical step is the determination of the initial model. As materials problems decrease in length scale, the electron microscope has proven to be a versatile and effective tool for studying many problems. However, study of complex bulk structures by electron diffraction has been hindered by the problem of dynamical diffraction. This phenomenon makes bulk electron diffraction very sensitive to specimen thickness, and expensive equipment such as aberration-corrected scatting transmission microscopes or elaborate methodology such as high resolution imaging combined with diffraction and simulation are often required to generate good starting structures.

The precession electron diffraction technique (PED), which has the ability to significantly reduce dynamical effects in diffraction patterns, has shown promise as being a "philosopher's stone" for bulk electron diffraction. However, a comprehensive understanding of its abilities and limitations is necessary before it can be put into widespread use as a standalone technique. This thesis aims to bridge the gaps in understanding and utilizing precession so that practical application might be realized.

Two new PED systems have been built, and optimal operating parameters have been elucidated. The role of lens aberrations is described in detail, and an alignment procedure is given that shows how to circumvent aberration in order to obtain high-quality patterns. Multislice simulation is used for investigating the errors inherent in precession, and is also used as a reference for comparison to simple models and to experimental PED data. General trends over a large sampling of parameter space are determined. In particular, we show that the primary reflection intensity errors occur near the transmitted beam condition and decay with increasing angle and decreasing specimen thickness. These errors, occurring at the lowest spatial frequencies, fortuitously coincide with reflections for which phases are easiest to determine via imaging methods. A general two-beam dynamical model based upon an existing approximate model is found to be fairly accurate across most experimental conditions, particularly where it is needed for providing a correction to distorted data. Finally, the practical structure solution procedure using PED is demonstrated for several model material systems.

Of the experiment parameters investigated, the cone semi-angle is found to be the most important (it should be as large as possible), followed closely by specimen thickness (thinner is better). Assuming good structure projection characteristics in the specimen, the thickness tractable by PED is extended to 40-50 nm without correction for complex oxides. With a forward calculation based upon the two-beam dynamical model (using known structure factors), usable specimen thickness can be extended past 150 nm. For *a priori* correction, using the squared amplitudes approximates the two-beam model for most thicknesses if the scattering from the structure adheres to psuedo-kinematical behavior. Practically, crystals up to 60 nm in thickness can now be processed by the precession methods developed in this thesis.

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Approved by

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