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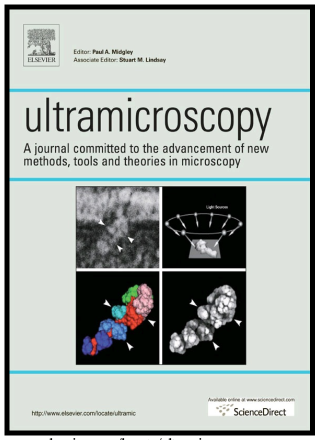

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A new method to detect and correct sample tilt in scanning transmission electron microscopy bright-field imaging

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Abstract

Important properties of functional materials, such as ferroelectric shifts and octahedral distortions, are associated with displacements of the positions of lighter atoms in the unit cell. Annular bright-field scanning transmission electron microscopy is a good experimental method for investigating such phenomena due to its ability to image light and heavy atoms simultaneously. To map atomic positions at the required accuracy precise angular alignment of the sample with the microscope optical axis is necessary, since misalignment (tilt) of the specimen contributes to errors in position measurements of lighter elements in annular bright-field imaging. In this paper it is shown that it is possible to detect tilt with the aid of images recorded using a central bright-field detector placed within the inner radius of the annular bright-field detector. For a probe focus near the middle of the specimen the central bright-field image becomes especially sensitive to tilt and we demonstrate experimentally that misalignment can be detected with a precision of less than a milliradian, as we also confirm in simulation. Coma in the probe, an aberration that can be misidentified as tilt of the specimen, is also investigated and it is shown how the effects of coma and tilt can be differentiated. The effects of tilt may be offset to a large extent by shifting the diffraction plane detector an amount equivalent to the specimen tilt and we provide an experimental proof of principle of this using a segmented detector system.

Keywords: Scanning transmission electron microscopy (STEM); annular bright-field (ABF) imaging; central bright-field (CBF) imaging; sample tilt

1. Introduction

Ferroelectric properties [1, 2] and octahedral distortions in perovskites [3, 4] are examples of technologically important material properties that can be inferred by precisely mapping picometer scale shifts in the positions of lighter atoms. Annular bright-field (ABF) imaging in atomic resolution scanning transmission electron microscopy (STEM) is a technique well suited for this purpose since it allows for simultaneous and robust imaging of both light and heavy atomic columns [5, 6]. Previous studies have shown that ABF is a good method of imaging columns of oxygen [5], lithium [7, 8] and even hydrogen [9, 10] atoms.

Precise position measurement in ABF is complicated by the fact that the column position measured from the ABF image will be laterally shifted from its true position for a small angular misalignment (tilt) of the sample axis with respect to the optical axis of the microscope. This was noted in the first theoretical studies of the technique [6] and has recently been explored in detail [11]. The observed shift is different for light and heavy elements, meaning that careful alignment of the specimen with the optical axis is necessary for very high precision measurements of relative atomic positions.

Careful alignment of a specimen requires a technique that is sensitive to tilt, such as position averaged convergent beam electron diffraction (PACBED) [12], which can determine sample tilts with a precision of about a milliradian. However this accuracy is diminished if a probe forming aperture with convergence semi-angle greater than about 20 mrad is used – exactly the kind of imaging setup necessary to optimize precise column position measurements using ABF imaging. Ideally a probe forming aperture with a smaller convergence semi-angle would be used to quantify specimen alignment and then a probe forming aperture with larger semi-convergence angle would be used to form STEM images but the process of changing the probe forming aperture generally in itself leads to some realignment of the optical axis and hence to specimen tilt.

In this paper we explain the underlying dynamics of why the influence of tilt of the specimen on the apparent position of light and heavy elements in a STEM ABF image is different. We introduce a method whereby specimen tilt can be determined from the STEM images themselves. This method can determine the tilt of a specimen to within a milliradian. We do this by placing a detector in the central part of the bright-field region that is not already occupied by the ABF detector – this is the central bright-field (CBF) detector. STEM images recorded using a very small CBF detector are reciprocal to images from bright-field conventional transmission electron microscopy (CTEM) [13], a technique that has been identified as being very sensitive to tilt [14], so we would expect STEM

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