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X-ray absorption in pillar shaped transmission electron microscopy specimens

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ABSTRACT

The dependence of the X-ray absorption on the position in a pillar shaped transmission electron microscopy specimen is modeled for X-ray analysis with single and multiple detector configurations and for different pillar orientations relative to the detectors. Universal curves, applicable to any pillar diameter, are derived for the relative intensities between weak and medium or strongly absorbed X-ray emission. For the configuration as used in 360° X-ray tomography, the absorption correction for weak and medium absorbed X-rays is shown to be nearly constant along the pillar diameter. Absorption effects in pillars are about a factor 3 less important than in planar specimens with thickness equal to the pillar diameter. A practical approach for the absorption correction in pillar shaped samples is proposed and its limitations discussed. The modeled absorption dependences are verified experimentally for pillars with HfO_2 and SiGe stacks.

1. Introduction

In thin transmission electron microscopy (TEM) specimens quantification of X-ray analysis is generally done with the Cliff-Lorimer method neglecting X-ray absorption in the lamellae [1]. This assumption is valid for X-ray lines with similar energies but can, even for specimens thinner than 100 nm, lead to appreciable error when low and high energy peaks are combined for the quantification [2–6]. These considerations become more important for X-ray tomography using pillar shaped specimens that often have diameters larger than the thickness of standard planar TEM specimens. A methodology to correct for X-ray absorption in 3D STEM-EDS tomography is recently discussed by Burdet et al. [7] based on a voxel-by-voxel calculation of the variation of the absorption along the X-ray track towards the detectors. The procedure is shown to be essential to correctly reconstruct the O and C distribution in core/shell nanowires with diameters on the order of 200 nm. Slater et al. [8] consider the maximum X-ray path length as criterion to decide whether the variation of the X-ray signal with tilt fulfills the projection requirement for tomography reconstruction in AgAu nanoparticles. They showed that for nanoparticles of only 40 nm diameter, this condition is still reached for the Au M line at 2.1 keV but not for the lower energy O K and C K lines. In the EDS reconstruction of II-VI multishell ZnTe/CdTe nanowires (diameter <100 nm) by discrete tomography using the a prior knowledge of the symmetry of the wires, the absorption of O and Mg is estimated to be less than 10% and taken into account for these elements in the ζ-factor quantification procedure [9]. To include a full nano-electronic device with gate and contact structures in a pillar TEM sample requires, also for advanced technologies, diameters exceeding 100 nm [10,11]. Such structures typically consist of a wide range of materials among which light elements that show strong X-ray absorption. Furthermore, in future technologies the active devices themselves also take cylindrical nanowires shapes [10,12-14], therefore further requiring the need to understand the absorption effects in pillar shaped morphologies.

Modern energy dispersive X-ray spectroscopy (EDS) systems are based on window-less Si drift detectors (SDD) that allow high count rates and have good sensitivity for low energy X-rays. Detection efficiency is further improved by increasing the X-ray collection angle by increasing the detector diameter and the use of multiple (2 or 4) detectors around the sample [15–17]. The latter can, depending on specimen and sample holder configuration, lead to important shadowing effects of the signal towards the different detectors [8,18–22]. In the 4-detectors geometry, the angular dependence of the shadowing for single tilt tomography holders with maximum tilt angles of ±70° shows a reduction of the X-ray intensities at 0° tilt to about 25% compared to the largest tilt angles [8,19,20]. As the signal shadowing is occurring by almost complete absorption in the heavy metal parts of the holder, the relative in-

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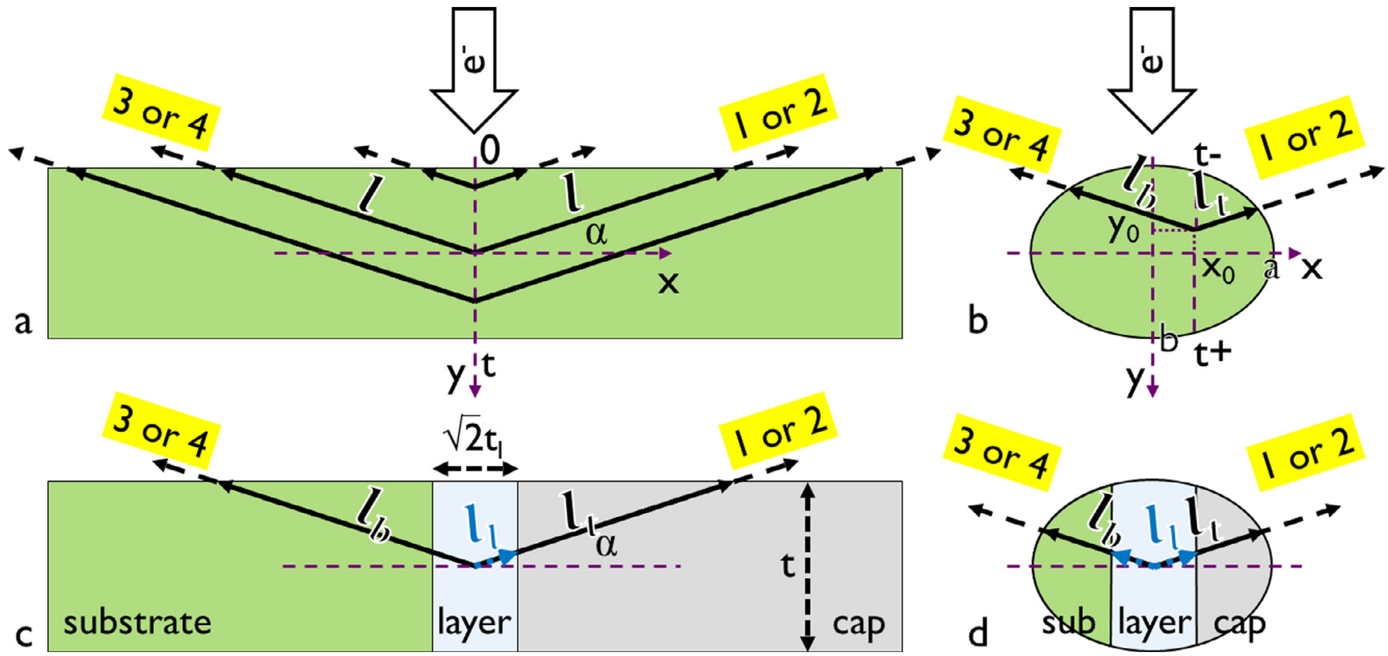

Fig. 1. Section in the plane of the electron beam and 2 opposite X-ray detectors for a planar (a) and a cylindrical pillar (b) specimen consisting of a single material in the configuration of Fig. 2d and a respectively. The electron beam is incident along the y direction. The X-ray path lengths in the middle of a layer with thickness t_l sandwiched between a substrate and cap material is shown for a planar specimen (c) and for a pillar (d). In the pillar the position of the layer shifts through the elliptical section while scanning along the diameter of the pillar.

tensities of high and low energy peaks is however not affected [8]. A time varied acquisition is proposed to compensate for the angular dependence of the X-ray intensities in such holders. In low-background double tilt holders the shadowing is reduced and the maximum count rates are obtained for 0° tilt [8,21] but in this type of holders the relative signal ratios of high and low energy peaks show an angular dependence due to different absorption strength of the signals in the Be-parts of the holder. This dependence can lead to incorrect quantification [8,21,22]. By a modified design of these holders the shadowing at 0° is strongly reduced [23]. With sufficiently high, free-standing pillar shaped specimens in 360° tomography holders shadowing effects are fully absent for all tilt angles. In that case the intensity ratio of high to low energy peaks is expected to be constant, unless absorption effects are induced by the structures in the pillar.

In this work the absorption effects in free standing pillar shaped specimens are modeled for different orientations of the pillar relative to the detectors in a 4-detectors EDS configuration. Universally applicable curves for the dependence along the pillar diameter of the relative intensities of weak and medium or strongly absorbed X-ray lines are derived. The practical application for absorption correction in pillar shaped specimens is discussed. The results are compared with experimental data acquired for pillars showing strong and medium absorbing effects, i.e. stacks with HfO_2 and SiGe respectively. The modeling can directly be extended to dual or single detector configurations as used in other types of microscopes.

2. Sample shape dependence of X-ray absorption

In plane specimens (Fig. 1a) the X-ray intensity $I(y)$ in the direction of the detector will be attenuated by absorption as can be described by the Lambert–Beer law [1]:

$$I(y) = I_0 e^{-l(y)/\lambda} \quad (1)$$

with $l(y) = y/\sin\alpha$ the X-ray absorption path length through the sample from the emission point towards the detector, α the take-off angle and λ the X-ray mean free path which can be calculated for a given matrix and X-ray energy from the tabulated X-ray mass attenuation coefficients μ/ρ [24–27] and the density ρ as:

$$\lambda = ((\mu/\rho)\cdot\rho)^{-1} \quad (2)$$

Integration over the specimen thickness t yields:

$$I_t = \int_0^t I_0 e^{-l(y)/\lambda} dy = I_0 \lambda \sin\alpha \left(1 - e^{-t/(\lambda\ \sin\alpha)}\right) \quad (3)$$

For pillar samples the specimen section in the plane of the electron beam and two opposite detectors is in general an ellipse (Fig. 1b). Therefore the X-ray path lengths $l(x, y)$ are dependent on the position x, y in the section and are, except for $x = 0$, different for detectors on the left and right side. Hence the integration over the specimen thickness becomes:

$$I_t(x) = \int_{t-(x)}^{t+(x)} I_0 e^{-l(x,y)/\lambda} dy \quad (4)$$

The path length $l(x, y)$ at each position x_0, y_0 can be calculated by determining the intersection point of the straight line to the detector $y = a'x + b'$ and the ellipse $x^2/a^2 + y^2/b^2 = 1$. The parameters of the line are given by $a' = \tan\alpha$ and $b' = y_0 - \tan\alpha\ x_0$ with α the take-off angle of the detector.

The path lengths $l(x, y)$ and the integrated intensities $I_t(x)$ are calculated numerically for the different detectors in a 4-detector EDS configuration for pillar specimens either aligned across or rotated 45° relative to the TEM specimen holder (Fig. 2a and b). The case of the pillar aligned across the holder is symmetry-equivalent with the one for a pillar along the axis of the holder as is the case in 360° tomography holders (Fig. 2c). The detector numbering is taken from the user interface of the TEM system and consistent with the tilt dependence of the signals. For a detector with 26 mm^2 active area at a distance of 12 mm [8,18,21] the semi-opening angle is ~14°. The opening angle of the detector is not taken into account in the calculations, i.e. it is assumed that the center of the detector well represents the average over its area. As will be seen in the comparison with the experimental data this is not always