TEM of lattice bending in crystallized areas of anodized Ta-O films

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Abstract. In the present paper we study the crystals grown in thin, initially amorphous films of tantalum oxide, interesting as possible materials for dielectric (high-k) layers to be applied in microelectronics. Here we employ some straightforward procedures making use of extinction bend contours (that are often observed in crystallizing amorphous films) and obtain indications of strong internal lattice bending in the crystallized areas.

1. Introduction
Valuable features of TEM (transmission electron microscopy) bend contour techniques were outlined in early papers on real space/electron crystallography (e.g., [1]) later on incorporated to a certain extent by convergent beam methods. Here we employ some straightforward procedures making use of extinction bend contours that are often observed (especially for oxide [2] and chalcogenide films/layers) though scarcely exploited in TEM studies of materials. In the present paper we study the crystals, grown in thin, initially amorphous films of tantalum oxide. The tantalum oxide thin films arouse special interest as possible material for dielectric (high-k) layers to be applied in microelectronics.

2. Method
Amorphous Ta-O films prepared by anodization were crystallized upon heating or/and electron beam annealing. Bend contours (BC) were assigned indices (matching SAD with bright and dark fields) and used to calculate the crystal lattice bending ($\phi$) measuring distance between zone-axis patterns (L, Fig. 1) or between contours in the pair (N, Fig. 2 b). As a result there were determined integral and local magnitudes of lattice bending (in fact, gradients of crystal lattice orientation) accordingly. Besides there was examined a period of BC fine structure oscillations (K, Fig. 2 d) to determine the depth of crystal intergrowth in the amorphous film.

The magnitude of the internal lattice planes bending was calculated by the following formulas:

$$\phi_{loc} = \frac{\lambda}{Nd_{hkl} \pi} \times \frac{180^\circ}{\mu m}; \phi_{int} = \frac{\delta}{L} \times \frac{180^\circ}{\mu m}$$

Where $\lambda$ — electron wavelength, $d_{hkl}$ — interplanar distance, $\delta$ — angle between zone axes, corresponding to the zone axis patterns. For the thickness we used:
3. Results

The crystallized areas present small lath-shaped single crystals (length up to several micrometers, Fig. 1, 2) with regular zone axis patterns outgoing from larger crystals with irregular broad bend contours (indicating much smaller crystal bending). Most of them are surrounded by fine grained material resembling porous polycrystalline structure (the grain sizes ~0.05–0.2 μm). Predominant phase was identified as hexagonal Ta$_2$O$_5$.

![Figure 1](image1.png)

**Figure 1.** Whisker of Ta$_2$O$_5$ with indexed BC pattern (a), SAED from ZAPs (b, c), dark field image in (300) (d)

![Figure 2](image2.png)

**Figure 2.** Whisker of Ta$_2$O$_5$: bright field (a, b) and dark field (c, d). Scheme of the measurements for calculations
The main data for the crystal lattice bending are summarized in the graph (Fig. 3), which is in parallel with our earlier data for some other substances: the thinner the crystal is, the larger is the crystal lattice bending [3]. The maximal lattice bending is around 190°/μm in fine grained areas and 80°/μm in whiskers.

Several earlier examples of individual crystals growing in amorphous films with strong lattice bending were/have been reported (e.g., in [2] where such crystals were introduced with the term “transrotational”), with a set of facts proving that it is an internal bending of the crystal lattice planes (in flat crystals) rather than a case of a mere crystal bending. In particular in our case the foil surface relief is too flat (Fig. 4) to explain the magnitude of crystallographic misorientation in single crystals.

The fact that the data, obtained earlier for various substances and in this report (for different microstructures: poly- and single crystalline) is a strong indication of a universal mechanism for the internal lattice planes bending in crystallizing amorphous films.

Regular bend contour patterns of the similar kind (probably indicating strong internal lattice bending) have been recently published for crystals growing in amorphous ALD- Ta₂O₅ films [4, 5].

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