In-Situ Transmission Electron Microscopy of Hafnium Zirconium Oxide for Phase Identification in Memristor Devices

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Hafnium zirconium oxide (HZO) has attracted much attention for the development of memristive technologies essential for neuromorphic computing, which is based on ferroelectric switching behavior attributed to orthorhombic polar phase (OPP) formation. This phase is stabilized by thermal strain induced by electrode type and process conditions. However, due to the polymorphic nature of HZO, formation of other stable polycrystalline phases with monoclinic, tetragonal and cubic structure is also possible. The identification of the crystalline phases by standard techniques is a challenge due to the similarity of lattice parameters and symmetry along some orientations, making it difficult for insightful optimization of OPP formation in these devices.

Herein we performed in-situ transmission electron microscopy, combining electron energy loss spectroscopy (EELS) and position averaged convergent beam electron diffraction (PACBED) to directly examine amorphous to crystalline phase transformation in HZO films under rapid thermal annealing. In EELS the signatures to identify the different phases was investigated by a detailed examination of the fine structure near the O_k edge. In PACBED we combine dynamical diffraction simulations with neural network based machine learning (ML) to examine distinction between the different phases. A high resolution TEM image of the crystalline phase formation is presented in Fig. 1(a) along with an inset showing its digital fast Fourier transform after rapid heating to 700^{o}C (at 50^{o}C/sec). Figure 1(b) is an EELS profile comparing the O_K edge from an amorphous region (Blue) before and the crystalline region (Red) after the annealing operation. Noticeable shift in peak-B position and a reduction in the B/A peak ratio are evident. While conventional diffraction analysis confirmed the formation of OOP, application of ML was inconclusive. Further details on the interpretation of changes in the O_K edge fine structure based on EELS modeling, and challenges with the ML approach to electron diffraction data analysis will be presented,

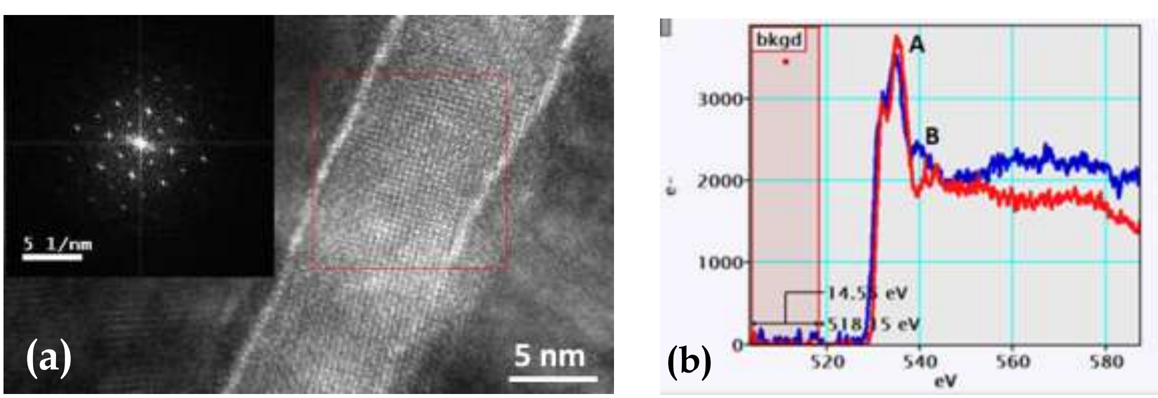

Fig1 (a) HRTEM image of a transformed region with inset showing the digital FFT of the image and (b) O_K edge EELS profiles showing changes in the near edge structure before (blue) and after heating (red).

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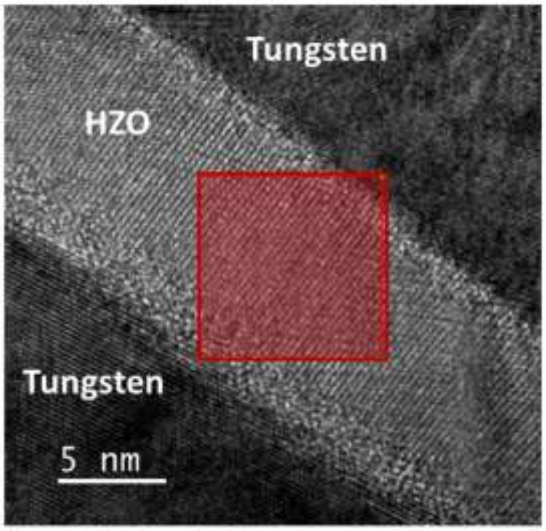
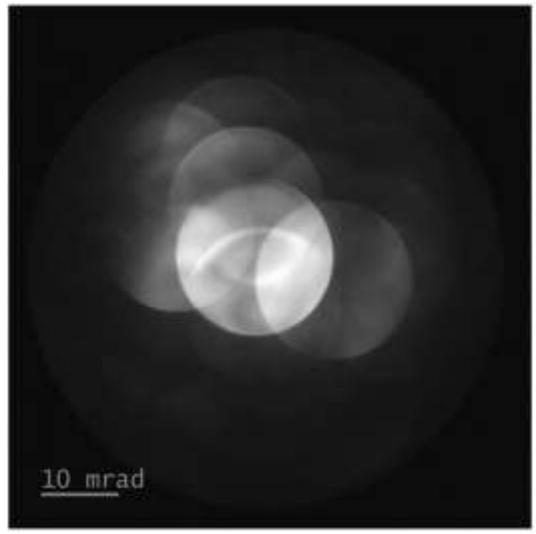
Supplementary Pages



HRTEM image of HZO grain within tungsten electrodes



PACBED pattern of the HZO grain from the region shaded in red