

Oxidation study of hafnium nanoparticles through grazing incidence x-ray diffraction

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Application of nanoparticles is of importance in various fields ranging from electronics to medicine. In our study we are interested to investigate metal oxide nanoparticles as building blocks of resistive memory switches a major candidate of future Non-Volatile Memories. HfO₂ thin films are among the strongest candidates for such memories [1] and investigation of nanoparticles made from this material could be of particular interest towards miniaturization of such devices. Furthermore the switching properties of such memories are affected by the crystallinity of the oxide films. In this context, towards application into resistive memory devices, we conduct a material study which involves fabrication and structure characterization of thin films composed from HfO₂ nanoparticles.

In the present work we examine the synthesis and characterization of HfO₂ nanoparticles obtained after oxidation of Hf nanoparticles that were fabricated by an inert gas condensation method. A vapor of Hf atoms metal is produced by dc sputtering of a hafnium target in a high pressure Ar atmosphere. The high pressure leads to aggregation of the atoms into particles. The particles are forced due to a pressure difference to enter a deposition chamber, where they soft land on the substrate. This method allows the fabrication of Hf nanoparticles with mean diameter about 5 nm.

The oxidation of the Hf nanoparticles into HfO₂ was performed either during the deposition by a beam of O₂ before they land on the substrate, or after the deposition with heating under O₂ flow to temperatures from 150°C to 600°C. The oxidized nanoparticles films were characterized by grazing incidence x-ray diffraction (GIXRD). This method is introduced as a direct, non-destructive, surface-sensitive technique for analysis of thin films. The method can be applied to polycrystalline thin films in order to determine the composition and structure of ultra-thin surface layers.

From the XRD spectra the determination of the conditions for the Hf nanoparticles oxidation was possible. The spectra reveals that oxidation of the Hf nanoparticles into HfO₂ is performed during the deposition of the nanoparticles under O₂ flow without application of temperature. Furthermore oxidation of the nanoparticles after their deposition in oxidizing atmosphere was also possible when the samples were heated to temperatures above 300°C. Also information about the changes in the polycrystalline structure of the HfO₂ nanoparticles film as a function of temperature was obtained. For low oxidation temperature the structure of the nanoparticles film consists of both monoclinic and tetragonal crystal lattices, while for higher oxidation temperatures only the tetragonal structure is observed. Furthermore the increase of the oxidation temperature leads to narrower peaks, which is indicative of the enhancement of the film crystallinity.

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References

[1] S. Long et al. ‘Quantum-size effects in hafnium-oxide resistive switching’, Appl. Phys. Lett. 102, 183505 (2013)

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