HfO₂/HfO_{2-x} Bilayer Structures for Multilevel Resistive Switching and Visualization of Oxygen Deficiencies by Electron Holography

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Resistive random access memory (RRAM) devices have attracted continuously intense research interests for non-volatile memory applications, thanks to their virtues like the low energy dissipation, the high switching speed and the good scalability etc. Hafnia (HfO₂) is one of the most intensively studied materials for RRAM, thanks to its compatibility with the Si-based complementary-metal-oxide-semiconductor (CMOS) technologies. Recently, many efforts focus on the multilevel resistive switching (RS) RRAM memristive devices and their rich RS dynamics, which open novel applications mainly on synaptic emulation for neuromorphic computing to link electronics with human brain functions. Meanwhile, unveiling the physical nature of the oxygen-deficient conductive filaments (CFs) that are responsible for the resistive switching of the HfO₂-based RRAM devices represents a challenging task due to the oxygen vacancy related defect nature and nanometer size of the CFs.

In this report, we presented a study on $Pt/HfO_2/HfO_{2-x}/TiN$ bilayer heterostructure, realized by reactive molecular beam epitaxy (RMBE). Such bilayer devices show enhanced resistive switching characteristics with multilevel behavior, indicating their potential as electronic synapses in future neuromorphic computing applications. Moreover, we demonstrate direct visualization and a study of physico-chemical properties of oxygen-deficient amorphous HfO_{2-x} in the bilayer structure by carrying out transmission electron microscopy electron holography as well as energy dispersive X-ray spectroscopy. Our results indicate the sensitivity of TEM EH to oxygen vacancy contents on a nanometer scale, which paves the way for future observation of CFs in HfO_2 films.