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Si Doped HfO_2 Anti-Ferroelectric Thin Films for Energy Storage and Solid State Cooling Applications

Authors : Faizan Ali, Dayu Zhou, Xiaohua Liu, Tony Schenk, Johannes Muller, Uwe Schroeder

Abstract : Recently, the ferroelectricity (FE) and anti-ferroelectricity (AFE) introduced in so-called 'high-k dielectric' HfO_2 material incorporated with various dopants (Si, Gd, Y, Sr, Gd, Al, and La, etc.), HfO_2-ZrO_2 solid-solution, Al or Si-doped $Hf_{0.5}Zr_{0.5}O_2$ and even undoped HfO_2 thin films. The origin of FE property was attributed to the formation of a non-centrosymmetric orthorhombic (o) phase of space group $Pbc2_1$. To the author's best knowledge, AFE property was observed only in HfO_2 doped with a certain amount of Si, Al, $Hf_xZr_{1-x}O_2$ ($0 \leq x < 0.5$), and in Si or Al-doped $Hf_{0.5}Zr_{0.5}O_2$. The origin of the anti-ferroelectric behavior is an electric field induced phase transition between the non-polar tetragonal (t) and the polar ferroelectric orthorhombic (o) phase. Compared with the significant amount of studies for the FE properties in the context of non-volatile memories, AFE properties of HfO_2-based and $Hf_xZr_{1-x}O_2$ (HZO) thin films have just received attention recently for energy-related applications such as electrocaloric cooling, pyroelectric energy harvesting, and electrostatic energy storage. In this work, energy storage and solid state cooling properties of Si-doped HfO_2 AFE thin films are investigated. Owing to the high field-induced polarization and slim double hysteresis, an extremely large Energy storage density (ESD) value of 61.2 J cm^{-3} is achieved at 4.5 MV cm^{-1} with high efficiency of ~65%. In addition, the ESD and efficiency exhibit robust thermal stability in 210-400 K temperature range and excellent endurance up to 10^9 times of charge/discharge cycling at a very high electric field of 4.0 MV cm^{-1}. Similarly, for solid-state cooling, the maximum adiabatic temperature change (

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