

論文の内容の要旨

論文題目 Impact of the Change in Temperature and Structural Distortion on the Spontaneous Polarization of Dielectric Interfaces and Ferroelectric Thin Films
(温度と構造歪みの変化が誘電体界面及び強誘電体薄膜中の自発分極に与える影響)

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In this study, the main motivation is to investigate how spontaneous polarization (P_s) can control the material properties of a material. For a material to possess P_s , the material must have a net polarization even in an absence of an external electric field. This study focuses on the P_s at dielectric interfaces and in ferroelectric (FE) thin films. In complementary metal-oxide-semiconductor (CMOS) and emerging non-volatile memory (NVM) memories, P_s is important because P_s in a gate dielectric determines the operating voltage of the device. This dissertation aims to find new methods to control the P_s at dielectric interfaces and in FE-thin films.

Due to the aggressive miniaturization of gate dielectrics, dielectric interfaces have become an essential part that determines the overall performance of a device. It is known that at a dielectric interface, a layer of dipole moments (also known as interface dipole layer) can be formed and causes a shift in the operating voltage of the device. So far, the magnitude (also known as interface dipole layer strength, φ_{dipole}) and the direction of an interface dipole layer are thought to be fixed with the choice of the material selection and the thermal annealing process. Hence, we extracted the temperature dependence of φ_{dipole} at several dielectric interfaces. We found that the φ_{dipole} is not fixed by the choice of materials selection and annealing process, but φ_{dipole} can also be controlled by the change in temperature. We also found that the magnitude of the temperature dependence can be universally explained using the concept of screening length.

The FE-thin film focused in this study is FE-HfO₂. FE-HfO₂ is a promising material that is a candidate to be the next material to be commercialized. Nevertheless, one of the remaining issues of FE-HfO₂ is the waking-up effect, which refers to the irreversible increase in the P_s during the initial cycles of electric field cycling. To date, the mechanism and the origin of the waking-up effect have already been established but not well discussed. Studies suggest that the origin of the waking-up effect is closely related to the phase transformation to the orthorhombic (O) phase¹ driven by electric field cycling; however, the waking-up effect and the phase transformation were not observed at the same scale. Therefore, the relationship between the waking-up effect and the phase transformation was not correctly represented. We clarified the origin of the waking-up effect by observing the two phenomena at the same scale. We found that the origin of the waking-up effect is the result of the phase transformation driven by electric field cycling.

Typically, it is unconventional for a phase transformation to take place without thermal annealing. Hence, the phase transformation must be governed by a phenomenon that has not yet been discussed. During structural characterization, we found an anomalous difference between the interplanar spacing (d_{spacing}) that is parallel and perpendicular to the surface of HfO₂ and attribute it to the structural distortion. We further investigated and clarified that structural distortion is a driving force that determines the amount of phase transformation to be driven by an electric field. Later, we also clarified that the anomalous structural distortion is originating from the rapid shrinkage in the volume of HfO₂ during the crystallization.

Normally, the thermodynamic stable phase in HfO₂ is regarded as the monoclinic (M) phase, but M-phase is non-FE. Although O-phase can become the stable phase in some circumstances, the phase transformation to the O-phase without thermal annealing is not kinetically feasible. We investigated the factors that accelerate the phase transformation to the O-phase without thermal annealing. We found that positive charges, the magnitude, and the time that the electric field was applied are important in the acceleration of the phase transformation to the O-phase.

To total, we found two new methods to control the P_s at dielectric interfaces and in FE-thin films; that is through the change in the temperature and the structural distortion, respectively.

¹ Orthorhombic phase is known to be the source of ferroelectricity in HfO₂.